

THE AMERICAN MINERALOGIST

JOURNAL OF THE MINERALOGICAL SOCIETY OF AMERICA

Vol. 21

JULY, 1936

No. 7

OPTICS AND STRUCTURE OF THREE-DIMENSIONAL SPHERULITES

H. W. MORSE AND J. D. H. DONNAY,
Stanford University and Johns Hopkins University.

TABLE OF CONTENTS

Introduction.....	392
Method of preparation.....	392
Description of some typical forms.....	394
Sheaves.....	395
Opening sheaves.....	396
Near-spheres.....	396
Mechanism of formation of spherulitic structures.....	397
Popoff's "Zweiblatt" in three-dimensional spherulites.....	401
Interference figure in parallel light.....	404
Optical distortion of the circular outline of a spherulite.....	407
The facts of observation.....	403
Canada balsam.....	403
Index liquids.....	409
Upper nicol only.....	409
Practical conclusion.....	410
Spherulitic structure theory.....	410
Various working hypotheses.....	411
Orientation of component crystals along spherulitic fibers.....	411
Analytical treatment in the case of uniaxial crystals.....	413
Discussion of equation (6).....	414
Remark.....	416
Biaxial crystals.....	416
Comparison between theory and experimental data.....	417
Form birefringence.....	413
Comparison between theory and experimental data.....	420
Lowering of indices in spherulitic fibers composed of aragonite crystals.....	422
Lowering of indices in spherulitic fibers composed of calcite crystals.....	424
Lowering of indices in fibers of SrCO_3 spherulites and BaCO_3 spherulites.....	424
Similar phenomena in fibrous minerals.....	424
Conclusion.....	425
Summary.....	425

INTRODUCTION (by J. D. H. D.)

The present paper gives the results of further investigation of artificial three-dimensional spherulites and related aggregates.¹ Financial help, received from the Penrose Fund of the Geological Society of America, to defray part of the expenses, and research facilities placed at our disposal by Stanford University, are gratefully acknowledged. Sincere thanks are tendered to Dr. G. Tunell for a critical reading of the manuscript.

Most of the laboratory work was done jointly by the two authors in the Summer of 1934. A first draft of the paper was then prepared. The manuscript, however, owing to alterations and additions, was not completed until March 1936, and Dr. Morse's illness at that time prevented him from seeing it in its final form. I, therefore, wish to take full responsibility for whatever errors or inaccuracies may be found in this paper.

Dr. H. W. Morse passed away on March 12, 1936 at the age of 63. May I be permitted to express at this place my deep admiration for his character as a man and as a scientist. His driving enthusiasm for art, literature, music, and drama, as well as for scientific research or professional duties, his serene philosophy, modesty, and kindness, made him a man of the highest type. Working with him has been a lasting source of inspiration, a thrilling experience, a great privilege. It is with a distressed emotion that I pay this tribute to his memory and express my profound gratitude for all I owe him.

METHOD OF PREPARATION

Artificial spherulites and related aggregates are prepared by allowing a concentrated solution of one salt to diffuse into a gel containing a dilute solution of another salt able to combine with the first by metathesis, and to form a compound of slight solubility. A great many substances can be grown in spherulitic form by this process; they are mentioned in our second paper. The present article, however, deals mostly with carbonates.

¹ Previous papers:

Morse, H. W., and Donnay, J. D. H., Calcite artificielle obtenue par diffusion dans un gel: *Bull. Soc. franç. Minér.*, vol. 54, pp. 19-23, 1931.

Morse, H. W., Warren, C. H., and Donnay, J. D. H., Artificial spherulites and related aggregates: *Am. Jour. Sci.*, vol. 23, pp. 421-39, 1932.

Morse, H. W., and Donnay, J. D. H., Spherulite optics: *Am. Jour. Sci.*, vol. 23, pp. 440-61, 1932.

Morse, H. W., Donnay, J. D. H., and Ott, Emil, Composition and structure of artificial spherulites: *Am. Jour. Sci.*, vol. 25, pp. 494-8, 1933.

Morse, H. W., and Donnay, J. D. H., Artificial spherulites: *Am. Mineral.*, vol. 18, pp. 66-7, 1933.

The gel used is gelatine in most preparations. Other gels have also been tried and proved satisfactory: agar, sodium silicate (water glass), fruit jellies, etc. Small spherulites have even been obtained in pure water by the same process of diffusion.

The best results are obtained with a commercial gelatine, previously washed and purified. However careful this process of purification, foreign ions seem to be unavoidable. The diffusion of barium chloride into a gelatine containing sodium carbonate usually yields a certain amount of optically positive spherulites, with low birefringence, together with negative spherulites, with high birefringence. Since only one substance should be expected from the double decomposition in progress, the other form may be thought to be due to impurities in the gelatine. This point was settled by an x-ray examination of the two forms present in different preparations; the negative one gave a powder picture identical with that of the expected carbonate, the positive one was proved to be a sulfate of the same metal (strontium, barium, lead). This explains the repeated observation that the positive form is more frequent in preparations obtained in a gel containing a higher percentage of gelatine. Gels ranging from 2 *per cent.* to 40 *per cent.* in gelatine content have been used; the higher the gelatine content, the more abundant were the positive spherulites, because of the larger amount of impurities in the gel.

An aqueous solution of gelatine, containing SO_4 ions, might be expected to precipitate, say, barium sulfate when a solution of barium chloride is allowed to diffuse into it. This was found not to be the case; the sulfate precipitates only in the presence of a carbonate in the gel (action of a protective colloid?).

The range of concentration of the two salts present may vary somewhat. We used 0.1N to 1.0N for the diffusing solution and 0.02N to 0.05N for the salt in the gel. The gel is allowed to set in a tube, placed vertically, and the concentrated solution is poured over it. The diffusion progresses downwards. The precipitation takes place in successive layers, obeying the law of rhythmic precipitation. Slight differences are observed in the form of the aggregates appearing in the lower layers (front of the diffusion) and in the upper layers (back of the diffusion). Differences in the rate of feeding of the growing aggregates by the diffusing substance probably account for this observation.

A great variety of forms are obtained in this manner. Their longest dimension is of the order of magnitude of 100μ . They can be washed out of the gelatine, dried, and mounted for optical examination under the microscope. They are always three-dimensional objects, sometimes perfect spheres. This point needs to be emphasized because the word "spherulite" is often used in the literature to designate two-dimensional

radiate crystallizations grown between glass slip and cover glass. Such spherulites actually are discs, in which the fibers radiate from the center like the spokes of a wheel. The latter have been extensively studied (Wallerant, Gaubert, Lacroix, Popoff) and it must be recognized that they afford much better material for investigation. An eminent example of this type of work is found in Boris Popoff's paper² on the modifications of malonamide and resorcine, whose optical properties are very carefully described and illustrated by exquisite plates.

Several papers have appeared in recent literature on the subject of three-dimensional spherulites. They seem to be of interest to a wide range of people. For instance, a botanist, A. Wieler, published an article on it last year.³ A book by C. J. Peddle on "Defects in glass" (kindly brought to my attention by Mr. W. J. Kirkpatrick) devotes a number of pages to descriptions of spherulitic crystallizations in industrial glasses,⁴ which will be of interest to all geologists acquainted with the paper by L. V. Pirsson dealing with spherulites in an artificial lava flow.⁵

DESCRIPTION OF SOME TYPICAL FORMS

In the preparation of artificial three-dimensional spherulites by diffusion and metathesis in gelatine and jellies, a large number of apparently different types of crystalline aggregates have been found, either alone or along with regular spheres. These aggregates assume the most capricious forms and, at first sight, seem to justify the view taken by those crystallographers who regard such unusual crystallizations as accidental. In his book *Trachten der Kristalle*, Tertsch writes:

We shall not consider here spherulites, ice-forms, crystal threads or hairs, sinter forms, and the like, since in these cases we have to deal with aggregates, whose external appearance (*habit* in the inexact sense of the term) is not a result of forces innate in the individual crystals, but a consequence of the more or less accidental aggregation of numerous individuals.

This statement is representative of a fairly common attitude towards aggregates.

Although it may possibly be true that some of the forms mentioned by Tertsch actually are accidental in origin, careful observation of our preparations shows convincingly that most, if not all, of our artificial aggregates belong to a well defined type of crystallization. The latter is

² Popoff, Boris, Sphärolithenbau und Strahlungskristallisation: *Latv. Farm. Žurn.*, 1934, pp. 1-48, February 1934.

³ *Kolloid-Zeitschrift*, vol. 70, p. 79, 1935.

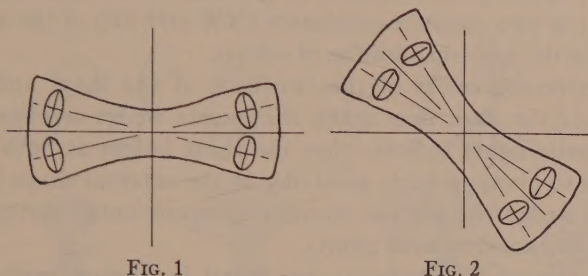
⁴ Published by Glass Publications, Ltd., Talbot House, Arundel Street, Strand, London W.C.2. 1927,

⁵ *Am. Jour. Sci.*, (4), vol. 30, p. 97, 1910.

characterized by sub-radiate to radiate arrangement of fibrous units whose component particles have the triperiodic crystalline structure.

The various forms we observed differ only from each other by their external shape, which is conditioned by the number of fibers entering its composition, that is to say by the more or less advanced stage of growth.

Sheaves.—Certain forms are bundles of fibers, somewhat divergent at both ends. We call them sheaves. They occur in preparations of a large number of substances (barium and strontium carbonates, calcium oxalate, etc.).



FIGS. 1-2. Behavior of a sheaf between crossed nicols. Sheaf in parallel position with respect to the nicols, in Fig. 1; sheaf at 45° to the nicols, in Fig. 2.

This is one of the first steps toward a radiate structure. When such an aggregate is examined between crossed nicols, a broad dark cross appears when the length of the sheaf is placed parallel to the vibration direction of one of the nicols; the four quadrants are faintly visible, they compensate as do negative spherulites (in the case of carbonates, *e.g.*), interference colors going up in two opposite quadrants and down in the other two quadrants. When the stage is rotated until the sheaf has its elongation at 45° to the nicols, the black cross disappears and so do the quadrants. The whole aggregate shows either addition or subtraction as the test plate is inserted. The sign of its elongation is negative, as should be expected from the preceding test in parallel position. This behavior is easily explained (Figs. 1 and 2).

When the sheaf lies in parallel position on the stage of the microscope (Fig. 1), the ellipses cut by a horizontal plane in the index ellipsoid of the negative fibers, although very slightly inclined on the sheaf axis, are inclined in opposite directions with respect to the ellipse of the test plate. In other words, the ellipses of the fibers have their major axes either almost parallel or almost perpendicular to the major axis of the ellipse of the test plate. Addition and subtraction, respectively, will take place, although the effect is not as clearly marked as it would be if these fiber ellipses were at 45° to the principal directions of the nicols

(as is the case in the quadrants of the *spherulitic figure*).⁶ Still it is sufficient to be discernible.

When the length of the sheaf lies at 45° to the nicols, the ellipses of the fibers have their major axes parallel or almost parallel to one of the axes of the test plate ellipse. Hence, either addition only (as in the case of the Fig. 2) or subtraction only will take place. This can also be visualized if the half sheaf be considered as a detached sector of a complete spherulite. In the first sketch (Fig. 1) the sheaf behaves like a horizontal diametral region (EW diameter) of the spherulitic figure (hence the cross and quadrants); in the second sketch (Fig. 2) the sheaf behaves like the central part of two opposite quadrants (NW and SE) of the spherulitic figure (hence the uniform addition of colors).

The interference colors of the two ends of the sheaf and also the movement of the dark lines when the quartz wedge is inserted (with monochromatic light) indicate that the sheaf bulges at both ends and permits one to obtain a fairly good idea of the external shape (the black lines in the wedge test are like moving "contour lines" giving approximately the thickness at each point).

Opening sheaves.—Such forms are found in barium carbonate and other substances, but are best shown in our preparations of calcium oxalate, where all degrees of "opening" of the sheaves may be seen.

Barium carbonate preparations in which widely opened sheaves occur also contain doublets and spherulites. The doublets look like two portions of spheres attached together.

The interference figure of the opened sheaves, obtained in parallel light, is very similar to that of a perfect spherulite. The more "open" the sheaf, the more striking the resemblance.

Near-spheres.—A widely opened sheaf gives the appearance of a sphere. Viewed under the microscope, it looks like a circle with a line drawn across a diameter, as it were, showing the region where the two growing ends of the sheaf have met.

In many cases, the result is a "doublet," in which the external shape of the two halves is close to spherical; these are viewed as two portions of circles united by a common chord, the size of the two portions being either the same or slightly different, the length of the chord being more or less different from a diameter.

It should be explicitly stated at this point that the words "opening sheaves" and the like, used in the foregoing descriptions, are not to be

⁶ The use of this term has been suggested in our third paper to designate the interference figure of a spherulite, obtained in parallel light and between crossed nicols. The figure consists of concentric rings of equal retardation and a dark cross; it is analogous to the convergent light figure exhibited by a uniaxial crystal cut perpendicularly to the optic axis; the law of spacing of the rings, however, is different.

taken literally. No actual "opening" takes place in the sense of fibers moving apart from one another; the apparent opening is due to further growth of new fibers at an angle to the old ones.

A series of sketches (Fig. 3) indicate the more common forms observed in the preparations.

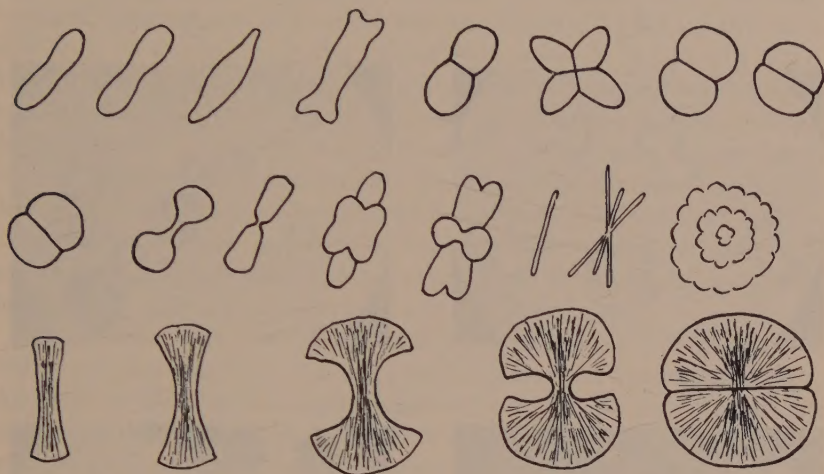


FIG. 3. Sketches of the more common forms observed in the preparations.

MECHANISM OF FORMATION OF SPHERULITIC STRUCTURES

When one tries to visualize the process of formation of spherulitic aggregates, the first idea that naturally comes to one's mind is that a spherulite must grow by simultaneous crystallization of fibers radiating from a common center in all directions with equal speed. Such a mechanism would obviously lead to a spherical shape of the aggregate at every moment during the growth. It is a "working hypothesis" in the sense that it will account for the phenomena observed on "perfect spherulites," that is to say those spherical aggregates which, in parallel light between crossed nicols, exhibit the "spherulitic figure" with ideal perfection. None of our observations can invalidate this hypothesis as a possible mechanism for such truly spherical aggregates. The explanation, however, cannot apply to the other forms described (sheaves, doublets, and the like). The growth of the latter would have to be explained by some entirely different process. Hence it would follow that the same chemical compound, which is found to assume different shapes in the same preparation, could almost indifferently adopt two (or more) different mechanisms of aggregate growth under apparently identical

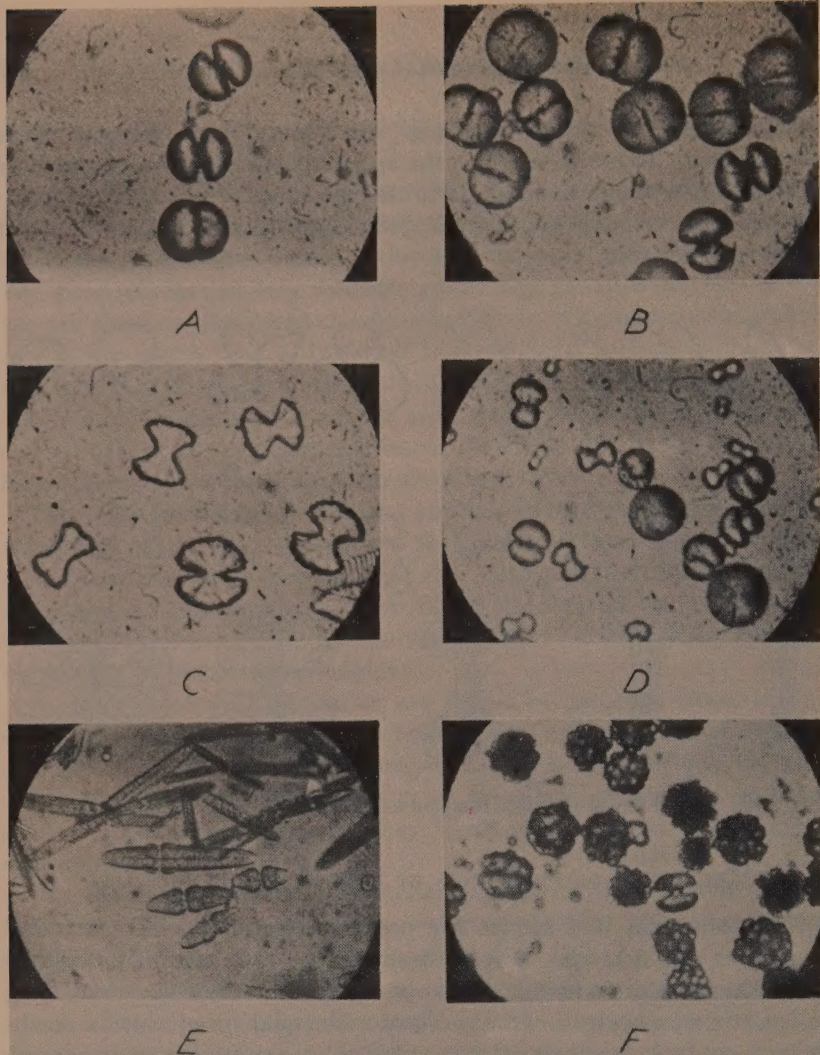


PLATE I

A—Strontium carbonate, widely opened sheaves or near-spheres. $\times 295$.

B—Strontium carbonate, same preparation as A. Mostly near-spheres and spheres showing the "waist line". Note that the size of the aggregate increases with the perfection of the sphere. $\times 295$.

C—Calcium oxalate forms, showing gradational transitions between sheaf and near-sphere. Note how further growth results in "opening" the sheaf like a double fan. Also note that the size of the aggregate increases as the "opening" progresses. One of our best photographs to substantiate our hypothesis as to the formation mechanism of such aggregates. $\times 295$.

D—Barium carbonate, similar forms. $\times 295$.

E—Barium carbonate, slender sheaves and doublets of several shapes, more or less elongated. The internal structure is fibrous and radiate. $\times 70$.

F—Barium carbonate forms, resembling mulberries. Although the external shape is rather irregular, only roughly spherical, the forms also belong to the general class of radiate aggregates. $\times 96$.

experimental conditions. This objection to a hypothesis of "isotropic growth" may not be irrefutable, yet it seems to offer a real difficulty.

Forms such as those found side by side in one and the same preparation of calcium oxalate (Fig. C, Plate I) can be interpreted as successive stages of spherulitic growth. The five aggregates shown in the photograph exhibit various degrees of "opening" of the sheaf and the more "opened" sheaves are larger than the less "opened" ones (in the ratio of 9 to 7, as measured on the two extreme cases in the photograph). Although these two facts combined will probably be considered sufficient by most geologists to warrant the interpretation of the various forms as successive stages, it is well to keep in mind just how much experimental evidence sustains the inference. We do not wish to revive the polemics that once divided "creationists" and "evolutionists," and shall simply state that, even if the inference be not fully substantiated, it at least is a working hypothesis leading to an unexpected, yet plausible, formation mechanism, valid for both sheaves and spheres.

In this conception the crystallization radiating from a center or nucleus must be considered as being possible in a limited solid angle only. We have no data on the angle of aperture of such a cone of radiating fibers, but it is obvious that a 360° aperture (crystallization radiating in all directions) is incompatible with the formation of sheaves. The latter is best explained by assuming an acute angle of aperture for the cone of possible radiating crystallization. The direction in which growth starts can reasonably be expected to be conditioned by the crystallographic properties of the substance. It should be the direction of fastest growth. This would account for the early stages of growth (fibers and fiber bundles). It must also be postulated, as a characteristic property of radiate crystallization, that every point at the surface of the growing cone of fibers can act as a new starting point for further radiating growth, the spatial extent of the latter being controlled by the possible angle of aperture of the cone, on the one hand, and by the mechanical obstruction of preëxisting fibers, on the other. As growth proceeds new fibers will appear to "branch out," that is to say will be formed at an angle to the older ones, and radiating from points already reached by the latter. The sheaf, hence, "opens" at both ends, in fan-like manner, until spherical shape is reached or, at least, approximated (doublets, near-spheres), when the feeding of the crystallizing aggregate gives out.

The shape of the intermediate stages will depend on the angle of aperture of the radiating crystallization. Figure 4 shows one-half of a meridional section of a growing sheaf, where the angle of aperture is arbitrarily taken equal to 50° for the cone of first radiation and equal to 14° for the successive radiations. Dashed curves indicate successive

outer surfaces. The velocity of growth is taken as constant (the same from all centers). When growth has proceeded sufficiently the "drooping" fibers issuing from the two ends of the sheaf will meet along a plane perpendicular to the axis of the sheaf (Fig. 4). The external appearance will undergo a radical change at this stage, passing as it does from a bundle to a near-sphere. The outer surface may be considered to be one

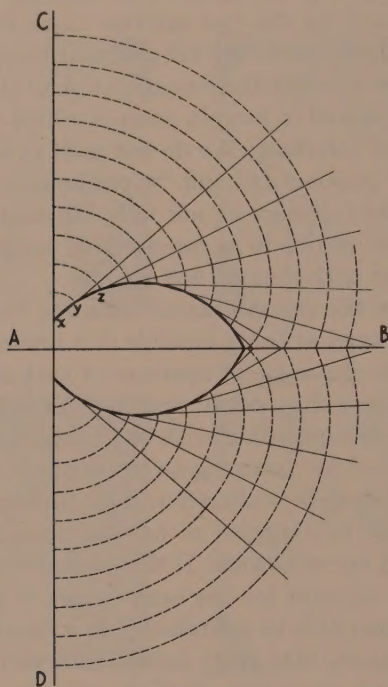


FIG. 4. Meridional section of a spherulite (one-half only) explaining the hypothesis as to its formation mechanism.

of revolution around the axis CD of the sheaf. It never becomes rigorously spherical, although it can approximate a sphere very closely. The reason for this is well shown in Fig. 4; the outer boundary, in the meridional section, is a curve composed of "raccording" arcs of circles with decreasing radii from the "polar regions" (at both ends of the axis of revolution) to the "equator," where the curve cusps in. The revolution surface must therefore show an equatorial constriction, which gradually becomes smoothed out (less and less reëntrant) as growth goes on. This feature has been repeatedly observed on near-spheres in our preparations. It was so striking that we used to refer to it as "the waist

line," in informal conversation, long before conceiving any possible theoretical explanation for it.

Will the center of a completed spherulite enclose a hollow region "walled in" by the fibers coming from both ends of the sheaf and meeting in the "waist line"? This would appear to be a logical consequence of our hypothesis. The shape of such a cavity, if it does exist, is easily visualized. The successive centers of radiation X, Y, Z, . . . (Fig. 4) are immediately seen to lie on an arc of a circle, as a necessary consequence of the mode of construction. This holds true for both the upper and the lower half of the sheaf. The lower half is the mirror-image of the upper in the equatorial plane AB. Hence the two arcs, upper and lower, have equal radii but different centers, and enclose a spindle-shaped area (shown in the meridional section, Fig. 4). Revolution of the latter around the axis CD of the sheaf generates the required surface of the cavity, which would look like the mould of a doubly umbilicated disc tapering off bluntly on its edge.

We have no experimental evidence in support of the existence of such a central cavity. If it does actually occur it must be small as compared with the size of the encasing spherulite, since in the vast majority of cases the spherulitic figure (parallel light, crossed nicols) shows no irregularity in the central region. It is more likely that the cavity is filled in by later growth. The example of calcium oxalate is suggestive in that respect since it shows (Fig. C, Plate I) that the empty space between the opening ends of the sheaf becomes narrower as growth proceeds.

POPOFF'S "ZWEIBLATT" IN THREE-DIMENSIONAL SPHERULITES

Since such transverse fibers as would be required to fill the hypothetical central cavity are not seen in the early stages (fiber bundles) they must either start growing later or else grow more slowly than the fibers parallel and sub-parallel to the axis of the sheaf. The idea of a difference in the velocity of growth is in accord with the crystallographic properties of a substance. It furthermore agrees with the experimental results obtained by B. Popoff in some two-dimensional spherulites. Popoff describes, and gives beautiful photographs of, a central region having the shape of a double leaf, which he calls "Zweiblatt."

The Zweiblatt is of course a plane figure, since the preparation is grown between glass slip and cover glass. Popoff calls attention to this fact and suggests that the central body may have a surface of revolution, so that the "double leaf" (Zweiblatt) seen in his preparations would be the meridional section of a "double bud" (doppeltknospenförmige Gestalt). The double bud is conceived as generated by the revolution

of the double leaf around its long axis (AB, Fig. 5). Notice that the surface generated by the revolution of the double leaf around an axis CD, perpendicular to the length of the leaves, is a doubly umbilicated disc similar (although not identical) to the one we discussed above as a consequence of our hypothesis of a limited, constant angle of possible radiating crystallization.

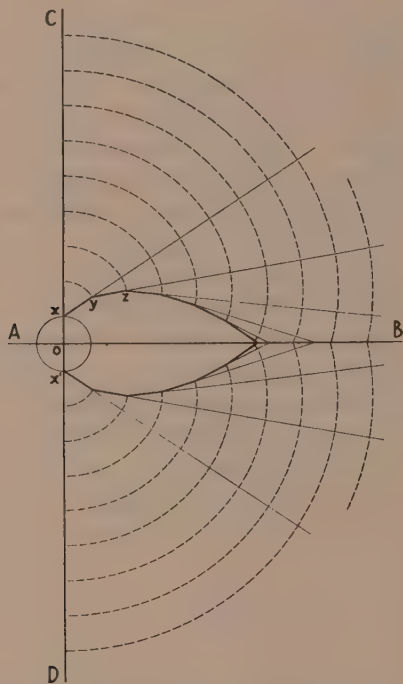
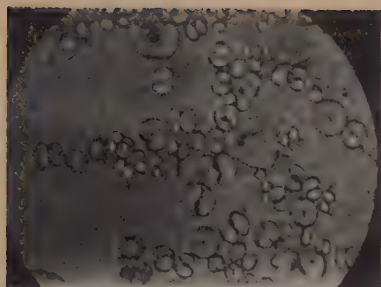
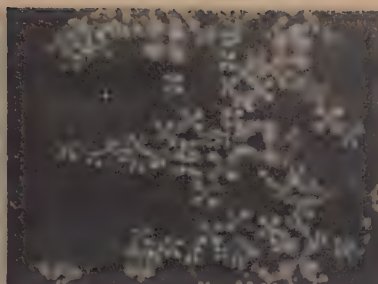


FIG. 5. Popoff's construction of the "Zweiblatt" curve (one-half only).

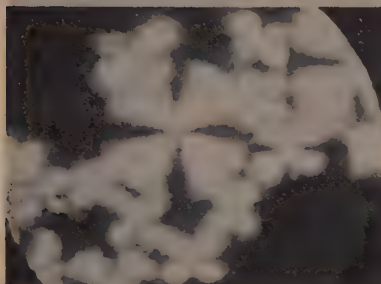
Popoff's elegant explanation of the shape of his Zweiblatt rests on the following assumptions: (1) The nucleus of the spherulite first grows in all directions with equal speed v_1 until a sphere is formed; the radius of this sphere may be extremely small but cannot be zero, otherwise the construction of the Zweiblatt curve (see above) would fail. (2) Two points, located at the extremities XX' of a privileged diameter of the initial sphere, then act as center points for radiating crystallization, which proceeds at a speed v_2 , slightly greater than v_1 , while the initial sphere keeps on growing radially at speed v_1 . The crystallographic orientation of the nucleus determines the diameter XX' as that of fastest growth. (3) Every point reached by radiating crystallization is



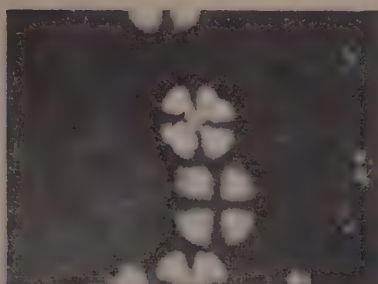
A



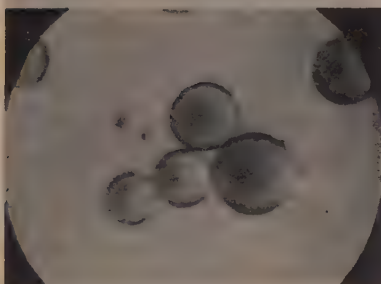
B



C



D



E



F

PLATE II

A—Calcium carbonate forms, ranging from more or less opened doublets to near-spheres and spheres. Note how the "waist line" persists through all stages; some spheres, otherwise perfect, show it as a diameter. Notice one hollow radiating aggregate (such forms have been observed by Cross in natural spherulites). $\times 167$.

B—Same as A, crossed nicols, parallel light. See gradations between perfect spherulitic figure and interference figures shown by doublets.

C—Calcium carbonate preparation containing occasional fairly regular spherulites, but mostly near-spheres and doublets. Note effect of external shape on interference figure and fibrous radiate structure, apparent from spherulitic cross. Crossed nicols, parallel light. $\times 167$.

D—Strontium sulfate. The near-sphere in center shows the "zweiblatt." Crossed nicols, parallel light. $\times 500$.

E—Barium carbonate. Perfect spherulites showing the optical distortion. Lower nicol only, tube somewhat lowered. $\times 96$.

F—Same as E, upper nicol only, tube somewhat lowered. The distortion is seen in opposite diameters, in photos E and F.

capable of acting as a new starting point for further radiation (this point was proved experimentally by Popoff). Radiating crystallization need not proceed in all directions, but only in a cone of "sufficient aperture." Popoff's construction yields a curve very similar to the observed Zweiblatt (see Fig. 5). It is as follows. From X as a center strike an arc with radius v_2 , from O as a center strike an arc with radius $2v_1$, the intersection of the two arcs is a point, Y, of the curve. Likewise Z is the intersection of an arc struck from Y as a center with radius v_2 and another arc from O with radius $3v_1$. And so on. Symmetrical branches of the curve will appear in the four quadrants and eventually meet on line AB.

From the construction just described, it appears that the central body of a three-dimensional spherulite is more likely to be a "disc" than the "double-bud" suggested by Popoff.

One of our photographs of strontium sulfate (Fig. D, Plate II) shows a doublet exhibiting the near-spherical shape, the equatorial constriction, and a central body. As far as we know, this is the first example of a central body in a three-dimensional spherulite. Compensation by means of the usual 1st order red plate indicates that the outer portion of the spherulite is optically positive, whereas the center is optically negative. It follows that the orientation of the crystalline particles along the fiber is different in the central body and in its encasing near-sphere. This difference of fiber orientation is known in two-dimensional spherulites, where it can be easily proved. In our case, however, we must reckon with another possibility, namely that the core be strontium carbonate and the shell strontium sulfate. The two salts, as has been explained above, have been found to occur together in the same preparation. We have been unable to rule out this last explanation; the diameter of the spherulite with a central body being 20 microns. We can only report the suggestive similarity of the central body shown in our photograph with Popoff's Zweiblatt.

We will now turn our attention to some of the optical phenomena displayed by spherulitic aggregates.

INTERFERENCE FIGURE IN PARALLEL LIGHT

As to the interference figure shown by the aggregates in parallel light between crossed nicols, it varies of course with the external shape, but the phenomenon is essentially the same as in a perfect spherulite. The "rings" of equal retardation are not as perfectly circular, but they are still visible in a great many cases. The "sign" of the figure is obtained as in a spherulite by examining the motion of the rings when a quartz wedge is inserted.

In a previous paper (*Spherulite optics*), we have shown that the movement of lines in the center of a spherulitic figure is the same as in the interference figure of a uniaxial crystal cut perpendicularly to the optic axis, and viewed in convergent light. This remark was sufficient to give an idea of the phenomenon in the case of perfect spheres. Since the phenomena are less plainly displayed in aggregates whose shapes depart sometimes widely from the spherical form, the analogy with the uniaxial convergent light figure becomes rather remote.

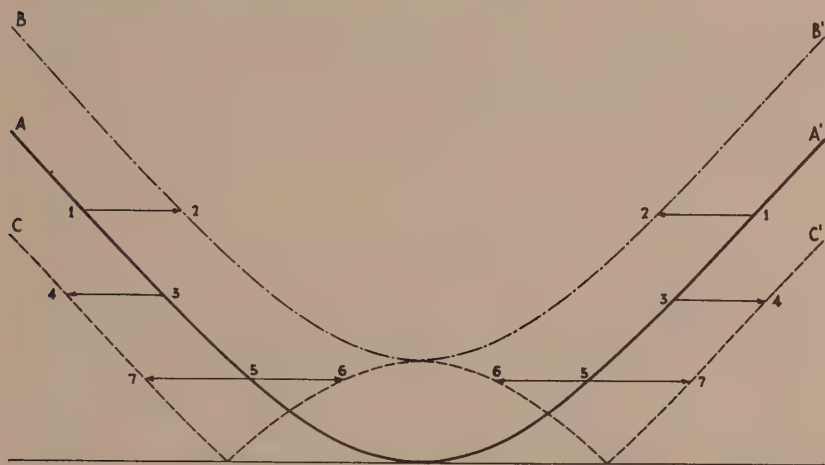


FIG. 6. Meridional section of the retardation surface in the interference figure (crossed nicols, convergent light) of a uniaxial crystal cut perpendicularly to the optic axis (curve AA'). The same, after inserting a test plate in additive position (curve BB'). The same, after inserting a test plate in subtractive position (curve CC'). Motion of isochromatic rings shown by arrows, when a quartz wedge is used instead of the test plate.

The retardation curve⁷ for the uniaxial figure is shown in figure 6. A uniaxial crystal is cut normally to the O.A. and examined in convergent light. The concentric circles are cut by a diametral line; the graph shows the retardation at each point along that line, *i.e.*, the value of the retardation (path-difference) is plotted against the distance from the center of the interference figure. The curve obtained has approximately the shape shown in Fig 6. The retardation increases from zero at the center of the figure, symmetrically on either side, as shown by

⁷ The equation of this curve can be found in our third paper, equation (13). Its derivation was based on the known theorem: the interference figure is similar in shape to the figure obtained on the upper face of the crystal slice if the light source is supposed to be a point on the lower face of the slice.

curve AA'. When a test plate is inserted in additive position, the curve of retardation becomes BB', since the retardation is increased at each point by the same amount, namely the retardation of the test plate. When the same test plate is used in subtractive position, the retardation is shown by the curve CC'. Note that retardations are always considered in absolute value so that there will now be a ring of zero retardation from which the retardation increases both outward and inward. The motion of the lines when a quartz wedge is inserted is shown by the arrows on the graph; in the case of addition, the ring numbered 1 shrinks into 2 (all rings shrink); in the case of subtraction, the outside rings expand (3, for instance, goes into 4), but the inside rings move both in and out (ring 5 contracts to 6 and expands to 7).

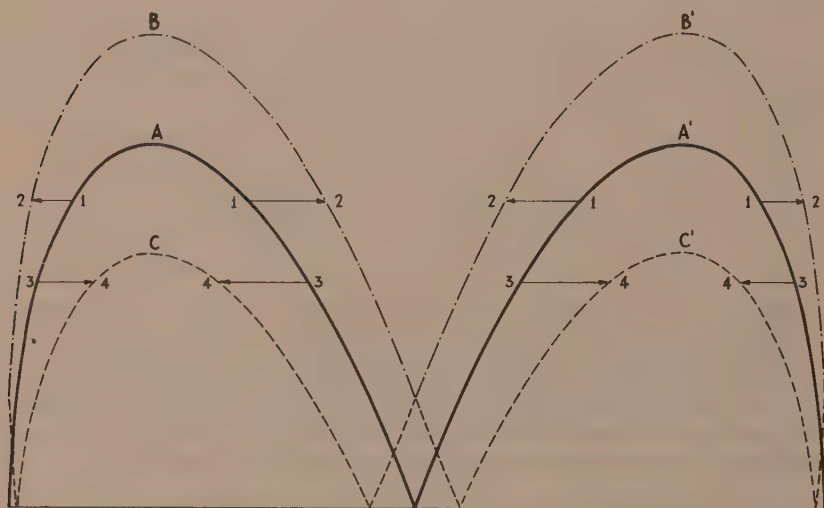


FIG. 7. Meridional section of the retardation surface in the spherulitic figure (crossed nicols, parallel light) of a perfect spherulite (curve AA'). The same, after inserting a test plate in additive position (curve BB'). The same, after inserting a test plate in subtractive position (curve CC'). Motion of isochromatic rings shown by arrows, when a quartz wedge is used instead of the test plate.

The retardation in a perfect (spherical) spherulite will now be considered (Fig. 7). The curve AA' shows the retardation across a diameter in a spherulite. The two maxima indicate the ring of maximum retardation (see third paper). When the wedge is inserted in additive position, the two concentric rings of retardation 1 move away from the ring of maximum retardation; when the wedge is used in subtractive position,

the two rings move towards the ring of maximum retardation (for instance, rings marked 3 go into 4). The inner rings (smaller than the ring of maximum retardation) shrink or expand like those of a uniaxial figure; the outer rings show the opposite effect.

If one keeps the above observation in mind, it is fairly easy to understand the figures shown by irregular (not spherical) aggregates, in which the line of maximum retardation is not a perfect circle and does not lie at exactly the same distance from the center as in a spherulite.

OPTICAL DISTORTION OF THE CIRCULAR OUTLINE OF A SPHERULITE

Perfect spherulites (perfect in the sense that they are very close to spherical), such as we have obtained in our artificial preparations, appear as true circles when viewed under the microscope in ordinary light.

In polarized light, however, certain spherulites show an appreciable amount of shading at the two ends of the NS (or EW) diameter (Figs. E and F, Plate II). This effect results in the appearance of two dark crescents giving the optical illusion of a shortening of the diameter that connects them. The outline of the perfect spherulite, in such cases, is no longer circular but elliptical. The optical effect is due to the radiate structure of the spherulite since the latter is the cause of the fact that the plane polarized light (vibrating NS, for instance) will be transmitted through the spherulite with different velocities along the NS and the EW diameters. In other words, the index of refraction effective at the ends of the EW diameter is different from that effective at the ends of the NS diameter. Depending upon the value of the refractive index of the mounting medium, the relief at the ends of the NS and EW diameters will be more or less different, hence the observed effect.

It is clear that the Becke test may give opposite results at the ends of the NS and EW diameters, if the index of the mounting medium is intermediate between the two indices of the spherulite effective at these two places. This has already been observed by Spangenberg⁸ for natural spherulites. This effect was thought to be susceptible of application to the measurement of the indices of the spherulitic substance by Spangenberg, who did not make any discrimination between the indices of the fiber and those of the substance constituting the fiber. The method, however, will only permit the determination of fiber indices; it can give no indication as to the indices of the substance entering the constitution of the fiber.

⁸ *Centralbl. Min.*, 1920, pp. 406-14.

The facts of observation.—Consider artificial spherulites of strontium carbonate. The identity of the spherulitic substance with strontianite has been proved by an earlier x-ray study (see fourth paper). Strontianite is slightly biaxial but can be considered uniaxial for our purpose; its indices (as given by Larsen) are: $n_e = 1.520$, $n_o = 1.667$. The fiber indices have been measured on splinters of the spherulites. The first measurements (previously reported) gave: $n_e = 1.516$, $n_o = 1.593$. The values of fiber indices apparently are apt to vary for, on repeating the measurements, we found: $n_e = 1.5095$, $n_o = 1.5955$. The following experiments, therefore, should be made on spherulites from the same crop, whose fiber indices have been previously determined. The spherulites are examined in different mounting media, in polarized light (lower nicol only, light vibrating NS).

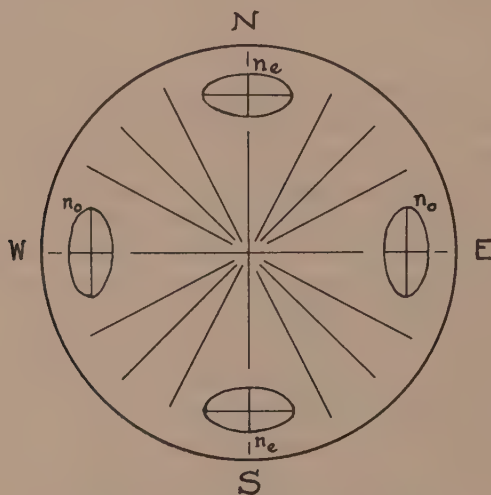


FIG. 8. Outline of a spherulite, as viewed under the microscope. Explanation of the optical distortion observed when a spherulite is embedded in a liquid whose refractive index is intermediate between the fiber indices.

Canada balsam.—Embedded in Canada balsam (index *ca.* 1.54), the spheres show the following effects.

When the tube of the microscope is raised, the Becke line goes in the spherulite at the ends of the EW diameter and out (*i.e.*, towards the medium) at the ends of the NS diameter. The ingoing line (EW) is followed by the dark crescents mentioned above. When the tube is lowered, the effect is reversed; the Becke line goes out EW and in NS, again followed by the dark crescent in the latter case.

The explanation of these observations is found in the following sketch

(Fig. 8), which shows the effective ellipses of the fibers of the spherulite. By "ellipse" we mean the ellipse cut through the ellipsoid of indices by a plane section parallel to the wave-front. The fibers are negative ("length fast"), hence the effective index, for light vibrating NS, is n_e at the ends of the NS diameter and n_o at the extremities of the EW diameter. Now the index of Canada balsam is higher than n_e and lower than n_o ; hence the observed effects.

That the fiber indices, not the crystal indices, are the important factor in the explanation must be proved, since in the case of Canada balsam the index of the mounting medium is comprised between the two crystal principal indices (n_e and n_o) as well as between the two fiber principal indices (n_e and n_o), and no conclusion can be drawn as to which are effective.

Index liquids.—A first liquid is chosen, whose index lies between the principal indices of the crystal, though not between those of the fiber. Let n be the index of the liquid. We have, in this case $n_e < n_e < n_o < n < n_o$.

The Becke test shows no difference in the behavior of the NS and the EW diameters. On raising the tube, the bright line goes out of the spherulite; on lowering the tube, the fringe goes in; and in both cases the motion of the line is concentric to the outline of the spherulite. There is a difference of relief, higher NS than EW, in our experiment because our liquid has an index equal to 1.62 and consequently $n - n_e > n - n_o$.

A second liquid is then used, whose index (designated here by n') is such that we have the relation $n_e < n' < n_e < n_o < n_o$. In this case, the index of the liquid lies in between the fiber indices, but not in between the crystal indices. If the fiber indices are the controlling factor, the difference in the Becke test according to the direction (either NS or EW) of the diameter should be observed here. This is found to be so. When the tube is raised, the Becke fringe goes out NS and in EW; the reverse appears when the tube is lowered. In both cases, the black crescents follow the bright line in its inward motion.

Upper nicol only.—Similar effects are obtained when only the upper nicol is used. Strontium carbonate spherulites, embedded in Canada balsam, show a Becke line moving towards the spherulite NS when the tube is raised, EW when it is lowered. This is the reverse of what has been observed when the lower nicol only is used.

The explanation is that light is polarized by the spherulite. Along the NS diameter, for instance, the ordinary light is polarized in two directions at right angles, one NS (parallel to the fiber axis), the other EW (normal to the fiber axis). The same is true for the EW diameter of the spherulite. The upper nicol only lets EW vibrations through; hence, n_o for the NS diameter and n_e for the EW diameter. It will be remembered

that the effective indices, with the lower nicol only, were n_e and n_o , respectively, for the NS and the EW diameters of the spherulite.

When other mounting media are used, similar observations can be made. With a liquid of index 1.66, for example, the effect is the same as with the lower nicol only, in the sense that the Becke test is concentric; the higher relief, however, is now EW ($n - n_e$) instead of NS.

Practical conclusion.—The fiber indices can be approximately determined by embedding the spherulite in various media. As long as the motion of the Becke fringe is different EW and NS, the index of the medium lies between the principal indices of the fiber. When the Becke test is concentric to the outline of the spherulite, the index of the medium is either higher or lower than both principal indices of the fiber.

Such tests can have no bearing on the determination of the indices of the crystals constituting the fibers.

SPHERULITIC STRUCTURE THEORY

The spherulitic figure, observed in parallel light between crossed nicols, is susceptible of a quantitative explanation (see previous paper on *Spherulite optics*). This explanation rests upon consideration of the splinter (fiber or fiber-bundle) as the unit of the radiate structure.

The value of the retardation R in a spherulite is here assumed to be directly proportional to the diameter $2r$ of the sphere; the ratio $R:2r$ is plotted for each ring (monochromatic light) against the radius of the ring. This yields the experimental curve. A theoretical curve is obtained from the assumption that the fiber-bundle (splinter) is a uniaxial entity with its own indices; the theoretical values of $R:2r$ are functions of these fiber indices only; the latter are measured by immersion methods and their values, introduced in the expression of the retardation, permit plotting of the theoretical curve, which is found to check with the experimental curve.

This treatment is the first step in the explanation of spherulitic structure. All the assumptions made in the theoretical treatment are justified by facts. The radiate structure is seen in crushed spherulites, which break into sectors; it is also observed between crossed nicols in parallel light, since it is the cause of the spherulitic dark cross. That the fiber-bundle should behave like a uniaxial entity, with its optic axis parallel with the elongation of the fibers, is justified by the fact that no third index can be measured across the fiber, so that both principal indices normal to the elongation of the fiber must be equal or sufficiently close to each other to warrant the assumption; the fiber structure, as revealed by x -ray studies, points to the same conclusion.⁹ The helicoidal arrange-

ment of crystal particles along the axis of the fiber, well known in two-dimensional spherulites, would give the same statistical average for the refractive index transverse to the fiber as a random arrangement in all directions but the fiber-length.

The treatment is quite satisfactory insofar as the fiber is considered the structural unit. The indices of the fiber, however, differ from those of the natural mineral of the same chemical composition. This discrepancy demands further explanation. The next step in the investigation should be the elucidation of the fiber structure.

Various working hypotheses.—The discrepancy between the fiber indices and the indices of the corresponding natural mineral could be accounted for by polymorphism. The fiber, in this case, would be a new modification of the known mineral. Such an hypothesis was considered. It has been disposed of by our *x*-ray study of artificial spherulites, which showed that the fiber is composed of the known modification.

Other explanations must now be considered. Alteration of the values of the refractive indices may be due to the orientation of the component crystals of the known mineral constituting the fiber. Since the presence of some interstitial material (water, gelatine?) has been ascertained in spherulites, form-birefringence (Form Doppelbrechung) is another factor that may be held responsible for the observed lowering of indices.¹⁰

ORIENTATION OF COMPONENT CRYSTALS ALONG SPHERULITIC FIBERS

The question of crystal orientation in spherulites has been studied by

⁹ Quoting Bragg, W. L.: "If a bundle of asbestos fibres is placed in the path of an *x*-ray beam, a pattern is obtained similar to that of an ordinary crystal rotation-photograph. Apparently the fibres all have one crystal axis along the fibre-length, while being oriented in all directions around this axis," (*Zeit. Krist.*, vol. 74, p. 254, 1930).

¹⁰ The values of the refractive indices of strontianite have been given, together with the values measured, by immersion methods, on fibers of strontium carbonate spherulites.

Other data are as follows.

For barium carbonate spherulites,

fiber indices:	$n_e=1.511$,	$n_o=1.615$,
also	$n_e=1.519$,	$n_o=1.615$;
witherite indices:	$n_e=1.529$,	$n_w=1.6765$.

For calcium carbonate spherulites,

fiber indices:	$n_e=1.485$,	$n_o=1.599$,
aragonite indices:	$n_e=1.530$,	$n_w=1.6825$,
calcite indices:	$n_e=1.486$,	$n_w=1.658$.

In the above list, strontianite, witherite, and aragonite are considered uniaxial since their index ellipsoid approximates an ellipsoid of revolution. The inaccuracy resulting from the approximation is less than the errors of measurements.

Jansen.¹¹ His x-ray results confirm the statement of Bragg, quoted above, and justify the assumption we had used in our theory of spherulite optics, *viz.*, that the fiber behaves like an optically uniaxial entity. Jansen finds that a spherulite fiber is made up of crystals, each of which has only one direction (always the same) fixed with respect to the fiber axis; and that, except for that restriction, the crystal may have any orientation (one degree of freedom, rotational). In the cases studied by Jansen, the crystal direction that lies along the fiber axis is a crystallographic axis of the crystal. A complete theory, however, should also consider other

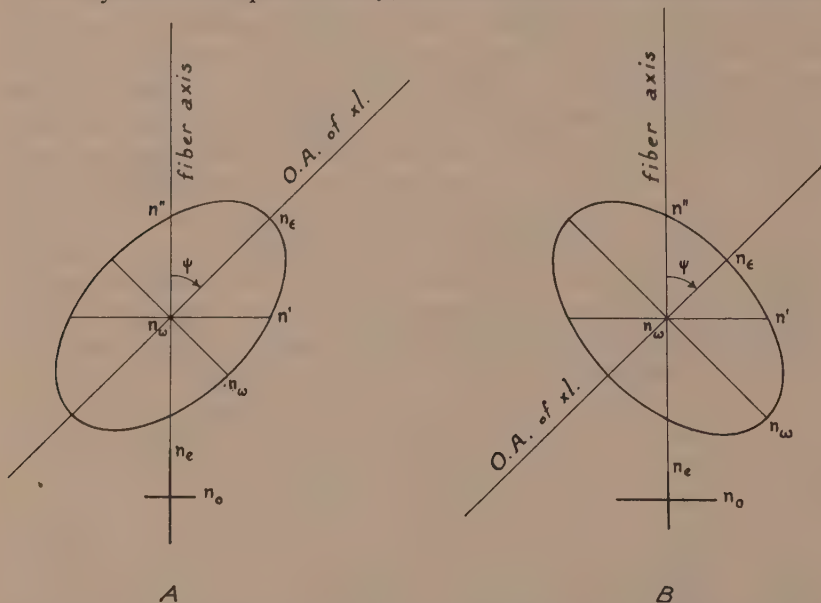


FIG. 9. Orientation of component crystals along the fiber axis. Positive crystal (A); negative crystal (B). Explanation of the relation between the fiber indices (n_e and n_o) and the crystal indices (n_e and n_o).

possible cases. From an optical standpoint, all cases would be covered by considering the problem as follows:

- (1) Uniaxial crystals—the optic axis of the crystal makes an angle ψ with the fiber axis, and ψ may vary from 0° to 90° .
- (2) Biaxial crystals—the direction of the fiber axis makes an angle ψ' with one of the optic axes and an angle ψ'' with the other. Both angles may vary from 0° to 90° . Optically the result is the same whether the fiber axis makes an angle ψ' with the first optic axis and an angle ψ''

¹¹ Jansen, Wilhem: Röntgenographische Untersuchung über die Kristallorientierung in Sphärolithen, *Zeit. Krist.*, vol. 85, pp. 239–70, 1933. (Bibliography of 26 titles.)

with the second optic axis, or an angle ψ'' with the first and an angle ψ' with the second. This is due to the symmetry of the triaxial ellipsoid.

Analytical treatment in the case of uniaxial crystals.—The problem is as follows. To find the indices n_e and n_o of a uniaxial fiber made up of uniaxial crystals with indices n_e and n_ω , the angle between the fiber axis and the optic axis of the crystal being ψ , an angle that varies from 0° to 90° .

Consider the cross-section of the ellipsoid of the component crystal by a plane passing through the fiber axis and the optic axis of the crystal (Fig. 9). Call n'' the index parallel to the fiber axis, represented by the radius vector of the "ellipse of section" making an angle ψ with the principal semi-axis n_e ; designate by n' the radius vector of the same ellipse at right angles to n'' . The equation of the ellipse in polar coordinates gives immediately

$$n' = \frac{n_\omega n_e}{\sqrt{n_e^2 \cos^2 \psi + n_\omega^2 \sin^2 \psi}}.$$

Using the same approximation as in our second paper (*Spherulite optics*), we may write instead of the above formula

$$n' = n_\omega \cos^2 \psi + n_e \sin^2 \psi \quad (1)$$

and we also have, in like manner,

$$n'' = n_e \cos^2 \psi + n_\omega \sin^2 \psi. \quad (2)$$

The indices of the fiber are

$$n_e = n'' \quad \text{and} \quad n_o = \frac{n' + n_\omega}{2},$$

or, after substitution,

$$n_e = n_e \cos^2 \psi + n_\omega \sin^2 \psi, \quad (3)$$

$$n_o = \frac{1}{2}(n_e \sin^2 \psi + n_\omega \cos^2 \psi + n_\omega). \quad (4)$$

The latter may be written as follows

$$n_o = \frac{n_e + n_\omega}{2} \sin^2 \psi + n_\omega \cos^2 \psi. \quad (5)$$

The *fiber birefringence* (or "spherulitic birefringence") is found to be, in terms of the *crystal birefringence* and the angle ψ ,

$$n_e - n_o = \frac{n_e - n_\omega}{2} (3 \cos^2 \psi - 1). \quad (6)$$

This expression is obtained by means of the equations (3) and (5). Note the occurrence of the second spherical harmonic ($3 \cos^2 \psi - 1$), encountered in other problems in physics.

The formulae numbered from (1) to (6) hold good for both positive and negative crystals (Figs. 9, A and 9, B, respectively); the birefringence of a positive crystal is positive ($n_e > n_o$), that of a negative crystal is negative ($n_e < n_o$).

Discussion of equation (6).—(1) For $\psi = 0$, $n_e - n_o = n_e - n_o$. The fiber birefringence is equal to the crystal birefringence, both in magnitude and in sign, when the direction of the crystal lying along the fiber axis is the optic axis. Example: quartzine, according to Correns and Nagelschmidt.¹²

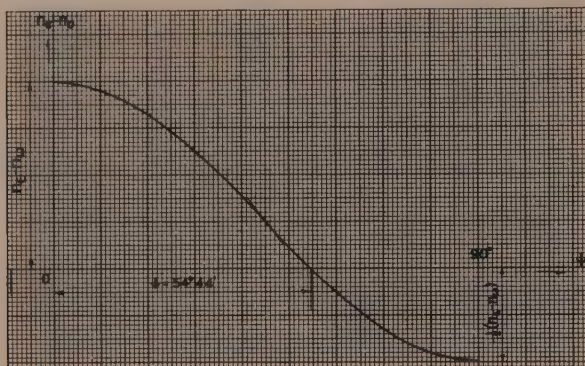


FIG. 10. Variation of the value of the fiber birefringence ($n_e - n_o$) with orientation of component crystals along fiber axis. The abscissa is the angle ψ between the axis of the fiber and the optic axis of the component crystal. The graph expresses equation (6), derived in the case of uniaxial crystals.

(2) For $\psi = 90^\circ$, $n_e - n_o = -\frac{1}{2}(n_e - n_o)$. The fiber birefringence is equal to one half the crystal birefringence and the sign is reversed when the optic axis of the constituent crystal is at right angle to the fiber axis. Example: chalcedony, where the fiber is made up of quartz crystals with either one of two horizontal crystallographic directions (*i.e.*, directions normal to the c -axis) oriented along the fiber length (x-ray results of Correns and Nagelschmidt).

(3) When the angle ψ varies from 0° to 90° , the fiber birefringence varies according to the law expressed by equation (6), passing from $(n_e - n_o)$ to $-\frac{1}{2}(n_e - n_o)$. There is a value of ψ for which the fiber bi-

¹² Correns, Carl W., and Nagelschmidt, Günter: Über Faserbau und optische Eigenschaften von Chalzedon, *Zeit. Krist.*, vol. 85, pp. 199-213, 1933.

refrindex is zero; it is found from the condition $3 \cos^2 \psi - 1 = 0$, which gives $\cos \psi = \frac{1}{3}\sqrt{3}$ and $\psi = 54^\circ 44'$. This is the crystallographic angle "cube:octahedron." The physical meaning of this is that it would be possible to build up optically isotropic fibers by means of birefringent crystals, uniaxial positive or negative, of any birefringence whatsoever, under the sole condition that the angle between the optic axis of each component crystal and the fiber axis be $54^\circ 44'$. This unexpected conclusion, however, is a consequence of the use of the approximate equation of the ellipse, introduced in equation (1). If the correct equation of the ellipse is used, the angle ψ for which the fiber birefringence is zero, has different values in positive and negative crystals; the two

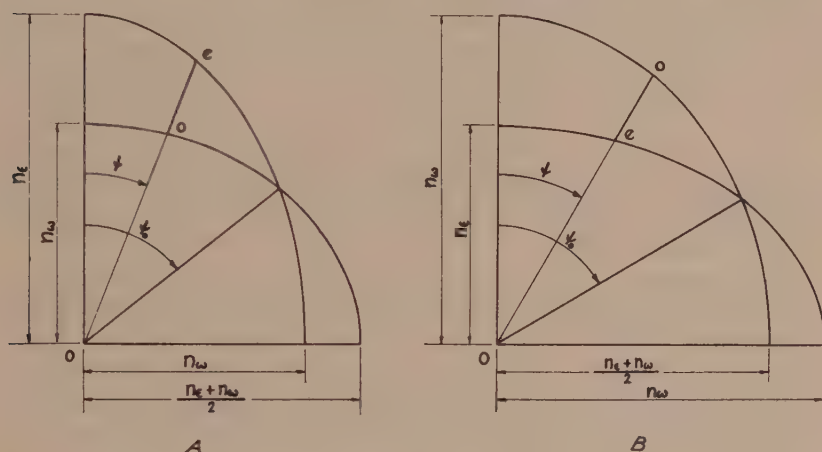


FIG. 11. A graphic way of finding the fiber indices (n_e and n_o) in terms of the crystal indices (n_e and n_o) and the angle ψ at which the optic axis of the component crystal is inclined to the fiber axis. Positive crystals (A); negative crystals (B).

values are astride $54^\circ 44'$, from which they depart but slightly in most cases. It must also be kept in mind that the present theory takes no heed of form-birefringence, the influence of which will be considered further on.

The curve expressed by equation (6) is plotted in rectangular coordinates on Fig. 10. It gives the value of the fiber birefringence ($n_e - n_o$) for any angle ψ comprised between 0° and 90° . The graph holds for positive and negative crystals; in the former case, the arrow on the ordinate axis indicates its positive direction; in the latter case, the same arrow marks its negative direction.

A graphic method of finding n_e and n_o , the fiber indices, in terms of n_e and n_o , the crystal indices, and ψ , the angle between the optic axis of

the crystal and the fiber axis, is shown in Fig. 11. Inspection of these graphs yields the following information:

In the case of positive crystals (Fig. 11, A), n_e can decrease from n_e to n_ω , but can never be smaller than n_ω ; n_o can increase from n_ω to $\frac{n_e + n_\omega}{2}$, but can never exceed the latter value. Likewise, in the case of negative crystals (Fig. 11, B): n_e can increase from n_e to n_ω but not any more, n_o can decrease from n_ω to $\frac{n_e + n_\omega}{2}$ but no further.

The values of n_e and n_o are not independent; their mutual dependence is shown by the graphs: when one increases, the other decreases.

As to fiber birefringence, $n_e - n_o$, the graphs show that it can retain the sign of the crystal birefringence $n_e - n_\omega$ and decrease from $(n_e - n_\omega)$ to 0, or can have the opposite sign and increase from 0 to $\frac{n_e - n_\omega}{2}$ in absolute value. Note that the fiber birefringence can never exceed the crystal birefringence when the sign is retained, also that the fiber birefringence cannot exceed one half of the crystal birefringence (in absolute value) when the sign is reversed.

The values $n_e - n_o = \frac{1}{2}(n_e - n_\omega)$ and $n_e - n_o = -\frac{1}{2}(n_e - n_\omega)$ occur for $\psi = 35^\circ 16'$ (the complement of the angle "cube:octahedron") and $\psi = 90^\circ$ respectively.

Remark.—A more exact solution of the determination of the fiber indices in terms of the crystal indices may easily be derived in the case of uniaxial crystals. In the preceding treatment, we have used the approximation generally found in text books on crystal optics.

For instance, for positive crystals:

$$n_e = \frac{n_\omega n_e}{\sqrt{n_e^2 \sin^2 \psi + n_\omega^2 \cos^2 \psi}} \quad \text{and} \quad n_o = \frac{2}{\pi} n_\omega F_1,$$

where F_1 is the complete elliptic integral of the 1st class,

$$F_1 = \int_0^{\pi/2} \frac{d\xi}{\Delta(\xi)}, \quad \text{with } \Delta(\xi) = \sqrt{1 - c^2 \sin^2 \xi}, \quad c^2 = \frac{n'^2 - n_\omega^2}{n'^2},$$

and

$$n' = \frac{n_\omega n_e}{\sqrt{n_e^2 \cos^2 \psi + n_\omega^2 \sin^2 \psi}}.$$

This solution, however, is only of mathematical interest because such accuracy is not required in our problem.

Biaxial crystals.—This case is somewhat more complicated than that of uniaxial crystals. We shall simply outline the procedure to be followed.

The radius vector determined in the triaxial ellipsoid by the direction of the fiber axis (making two given angles with the optic axes) is computed in terms of the three principal indices and the two angles ψ' and ψ'' . This is the value of the fiber index parallel with the fiber elongation, n_e . Then the two half-axes of the ellipse cut through the ellipsoid by a plane normal to the fiber axis are computed (the formulae solving this problem may be found in Fletcher's *Indicatrix*, for instance); the half-sum of these two indices is equal to the fiber index normal to the elongation of the fiber, n_o .

That such cases need be taken into consideration is evident since they are known in nature. The orientation of the crystal with respect to the fiber axis in fairly large spherulites of feldspars is given in the literature as the edge [100] along the fiber length. Since, in the cases studied in this paper, the index ellipsoid of the substances considered approximates an ellipsoid of revolution, there was no need for developing the theory of biaxial component crystals.

Comparison between theory and experimental data.—Consideration of the crystal orientation along the fiber is insufficient to account for the change in the values of the refractive indices apparently brought about by the aggregate structure.

The example of strontium carbonate spherulites (optically negative) will serve to demonstrate the lack of agreement between the theory and the measurements.

The indices of the strontianite crystals (negative) constituting the fiber are: $n_e = 1.520$, $n_w = 1.667$, as n_α is taken for n_e and $n_\beta = n_\gamma$ for n_w .

If n_e is assumed to be oriented along the fiber length, the fiber indices n_e and n_o should be, according to the above theory, $n_e = n_e = 1.520$, $n_o = n_w = 1.667$.

The measured values, one will recall, were $n_e = 1.5095 - 1.516$, $n_o = 1.5955 - 1.593$.

If we assume that n_e is perpendicular to the fiber axis, the fiber birefringence should be positive, with $n_e = n_w = 1.667$, $n_o = \frac{1}{2}(n_e + n_w) = 1.593$. Neither the indices nor the optic sign checks with the observations.

Can n_e be oriented at an angle ψ to the fiber axis that would account for the values of the measured indices? Substituting in the equation (6) the value of the strontianite birefringence (-0.147) for $n_e - n_w$ and that of the fiber birefringence (-0.086 or -0.077) for $n_e - n_o$, we get $\psi = 31^\circ 44'$ or $\psi = 34^\circ 18'$, according to the value adopted for the fiber birefringence. If we set ψ equal to $31^\circ 44'$ or $34^\circ 18'$ in equations (3) and (5), we reach respectively the following sets of values for the fiber indices: $n_e = 1.561$, $n_o = 1.647$; and $n_e = 1.567$, $n_o = 1.643$.

It is plain, therefore, that some other factor, besides the crystal

orientation along the fiber, must be taken into account in order to explain the lowering of the refractive indices. Let us consider the effect of form birefringence.

FORM BIREFRINGENCE

Form birefringence is an important case of accidental birefringence, accidental in the sense that it is not due to the anisotropy resulting from (non isometric) crystallinity.

It is well known that isotropic particles may build up an anisotropic fiber, which behaves either like a positive uniaxial or like a negative uniaxial entity (so-called "rod birefringence" and "plate birefringence" respectively). The formulae have been established by Wiener¹³ on the basis of the electromagnetic theory of light; they have been applied by colloid chemists (Ambronn and Frey, Wächtler¹⁴). Anisotropic particles (rods or plates) may, in like manner, build a fiber with a "form birefringence," which is algebraically added to the original birefringence of the fiber.

The condition for the existence of form birefringence is that the fiber be composed of two media, the particles of which must be smaller than the wave length of light. In their explanation of the structure of chalcodony spherulites, Correns and Nagelschmidt have postulated that this condition was satisfied. Our previous x-ray study of artificial spherulites (Morse, Donnay, Ott) gave us the order of magnitude of the crystalline particles in the aggregate—ranging from 10^{-3} to 10^{-5} cm. A number of these particles must be smaller than the wave length of light. As to the presence of another medium, interstitial between the crystal particles, it is to be expected that spherulites grown in gelatine will contain some interstitial medium (gelatine, water, or possibly air). Johnston, Merwin, and Williamson¹⁵ have reported the retention of as much as 4 per cent (by weight) of water by artificial spherulites of calcium carbonate. Moreover the results of specific gravity determinations on our artificial spherulites cannot be accounted for without the presence of some foreign medium, less dense than the substance of the spherulite. We therefore feel justified in considering form birefringence as a possible agent effective in the lowering of the indices.

It is difficult to give a quantitative treatment of the action of form birefringence in the present instance because neither the percentage amount nor the refractive index of the interstitial medium can be determined accurately.

¹³ *Abhandlungen d. Sächs. Ges. d. Wiss., math.-phys. Kl.*, 1912.

¹⁴ Wächtler, Maximilian, Über die Doppelbrechungserscheinungen in Kolloiden, *Fortschr. d. Min., Krist. u. Petr.*, vol. 12, pp. 119-192, 1927. Bibliography of 91 titles.

¹⁵ Johnston, J., Merwin, H. E., and Williamson, E. D., The several forms of calcium carbonate: *Am. Jour. Sci.*, (4) vol. 41, pp. 473-512, 1916.

The problem can be attacked in the following manner. First assume the orientation of the crystals with respect to the fiber axis; assumption to be guided by the habit of the crystal. Compute the indices of the fiber (n_e, n_o) in terms of the indices of the crystal (n_e, n_o) by means of the formulae (3) and (5), derived in the preceding section. Compare the "original birefringence" ($n_e - n_o$) with the "total birefringence" ($n_{\parallel} - n_{\perp}$), which is the difference of the measured indices of the fiber. This comparison will enable one to decide whether the "form birefringence" should be positive or negative. If it is found to be positive, for instance, the fiber must be a bundle of rods or needles; this affords a check on the assumption for the crystal orientation. Finally, solve Wiener's two equations for the *index of the interstitial medium* and its *percentage by volume*. Since we have two unknowns and two equations, there is no way of checking the results. Further progress will depend on devising an experimental technique of determining the volume percentage of the medium and its index of refraction.

The comparison of birefringences, mentioned in the above paragraph, is made by using the formula

$$B_{\text{total}} = B_{\text{original}} + B_{\text{form}}.$$

In this formula the original birefringence, B_{original} , of the fiber represents the birefringence that the fiber possesses owing to the anisotropy of its crystalline component particles; it has also been called "own birefringence" (Eigendoppelbrechung) by chemists; it is a function of the indices n_e and n_o of the component crystals; this function was designated by ($n_e - n_o$) in equation (6). The form birefringence, B_{form} , of the fiber, on the other hand, depends on the shape and arrangement of the component particles of the fiber, together with the presence of an interstitial medium; form birefringence exists even in a fiber composed of isotropic particles and an isotropic medium. The total fiber birefringence, B_{total} , should agree with the values derived from the measurements of the fiber indices; we shall designate it by ($n_{\parallel} - n_{\perp}$) in the following formulae, n_{\parallel} and n_{\perp} being the theoretical values that should agree with the measured fiber indices (given in footnote 7, where they were designated by n_e and n_o).

Wiener's formulae may be written as follows, in the case of positive form birefringence:—

$$n_{\parallel}^2 = (1 - x)n_e^2 + xy^2,$$

$$n_{\perp}^2 = y^2 \frac{(2 - x)n_o^2 + xy^2}{(2 - x)y^2 + xn_o^2},$$

in which n_{\parallel} and n_{\perp} are the indices of the splinter (fiber), n_e and n_o the indices of the fiber as resulting from the orientation of crystal particles,

y the index of the interstitial medium, and x the fraction of unit volume occupied by the interstitial medium (so that there is $100x$ per cent of medium by volume).

For negative form birefringence the formulae become, with the same notation,

$$n_{\parallel}^2 = \frac{n_e^2 y^2}{(1-x)y^2 + xn_e^2},$$

$$n_{\perp}^2 = (1-x)n_o^2 + xy^2.$$

Comparison between theory and experimental data.—Even when the action of form birefringence is taken into account, in addition to that of crystal orientation along the fiber, a comparison between the theoretical results and the measurements is disappointing.

One case only might be considered somewhat satisfactory, *viz.*, that of calcium carbonate spherulites. Here, however, the determination of which modification of calcium carbonate occurs in the spherulites presents an additional difficulty. The very careful work of Gibson, Wyckoff, and Merwin¹⁶ has shown that calcium carbonate can occur in spherulitic form in two modifications: vaterite *A* and vaterite *B*, which they have proved to be identical with calcite and μ -CaCO₃, respectively. They have not found any spherulites made of aragonite. The observations we have made on our spherulites of calcium carbonate do not fit the properties of either vaterite *A* (=calcite) or vaterite *B* (= μ -CaCO₃). The measured indices of the fiber are $n_{\parallel} = 1.485$, and $n_{\perp} = 1.599$; these values are decidedly too low for vaterite *B*; the value of n_{\parallel} is above the maximum value recorded for vaterite *A* by Johnston, Merwin, and Williamson (1916), although that of n_{\perp} falls within the range of measurements of these authors. The fibers have a negative elongation, like fibers of vaterite *A*. The cobalt nitrate test (Meigen reaction) unquestionably yielded the lilac coloration (supposed to be characteristic of aragonite), like vaterite *B*. An x -ray powder picture, kindly taken for us by Dr. Hoard, gave the calcite pattern, like vaterite *A*; on reexamination of the sample under the microscope, however, it was found that the spherulites, while retaining their external spherical form, had recrystallized into large, irregular areas of calcite; it is quite possible that this recrystallization may have taken place before the x -ray photograph was obtained, because several weeks elapsed before Dr. Hoard took his picture. At all events, the lowering of the refractive indices cannot be explained on the basis of form birefringence and crystal orientation, if the spherulites are made of calcite. The explanation is, on the other

¹⁶ Gibson, R. E., Wyckoff, R. W. G., and Merwin, H. E., Vaterite and μ -calcium carbonate: *Am. Jour. Sci.*, (5), vol. 10, pp. 325-333, 1925.

hand, fairly plausible if the spherulites are supposed to be composed of aragonite crystals, as will be shown presently.

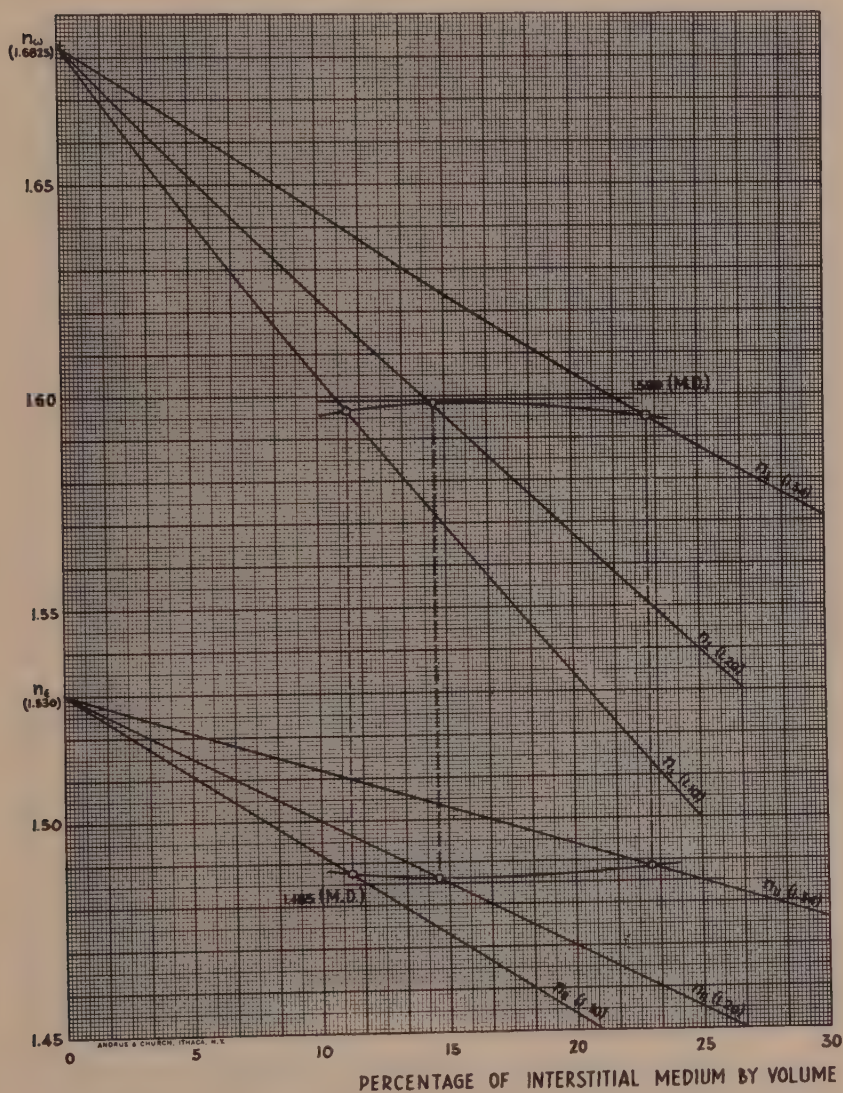


FIG. 12. Action of form birefringence in aragonite spherulites. Curves showing the lowering of the fiber indices (n_{\perp} and n_{\parallel}) with increasing percentage of interstitial medium. Three pairs of curves are drawn, the index of the medium being taken as 1.34, 1.20, and 1.10, respectively.

The measured values of the fiber indices, obtained by Johnston, Merwin, and Williamson (1916) for spherulites of vaterite *A* (= calcite), are satisfactorily accounted for by the theory.¹⁷

Lowering of indices in spherulitic fibers composed of aragonite crystals.—Let us apply the foregoing theory to spherulites made of aragonite crystals, forming fibers separated by a varying percentage of interstitial medium, for several values of the refractive index of the medium. Plotting the index against the percentage of interstitial medium, we shall obtain two families of curves, one for n_{\perp} and one for n_{\parallel} , the index of the medium changing from curve to curve in each family.

The indices of aragonite are: $n_{\alpha}=1.530$, $n_{\beta}=1.680$, and $n_{\gamma}=1.685$. We consider aragonite crystals to approximate uniaxial crystals with the following indices: $n_e=1.530$, $n_o=1.6825$. The acicular habit of aragonite (parallel with the *c*-axis) suggests the assumption that the fibers are made up of crystals oriented with their *c*-axis parallel to the fiber length. Since the orientation of the component crystals is random except for that one restriction, the fiber results in a uniaxial entity with the indices: $n_e=n_e=1.530$ and $n_o=n_o=1.6825$. The measured indices of the fiber are: $n_{\parallel}=1.485$ and $n_{\perp}=1.599$. The theory should account for the lowering of the indices from 1.530 to 1.485 and from 1.6825 to 1.599, respectively. The fiber birefringence, which *a priori* should be 0.153 (= original birefringence), only amounts to 0.114 (= total birefringence). These figures are absolute values, since these birefringences are negative. We, thus find that the form birefringence must be positive and equal to 0.039, in order to satisfy the relation $-0.114 = -0.153 + 0.039$. Two structures are possible: rods or plates, parallel to the fiber axis. Our assumption as to the orientation of crystals in the fiber is one of them (rods).

Plot the values of n_{\parallel} and n_{\perp} given by Wiener's formulae against *x*, the percentage by volume of the medium, for several values of the index *y* of the medium (1.34, 1.20, 1.10). The curves obtained are almost straight lines, showing only a slight concavity upwards (Fig. 12). Now the measured values of n_{\parallel} and n_{\perp} should be found on the same vertical (*i.e.*, for a given percentage of interstitial medium) on corresponding curves (*i.e.*, curves drawn for the same value of the index of the medium). The graph will show that the measured values of the fiber indices lie nearly on the same vertical for 15 *per cent.* by volume of an interstitial medium with index 1.20. Owing to the observed variability in the refractive indices of the fibers, the following figures may also be considered in satisfactory agreement with the measurements: 20 *per cent.* of a medium

¹⁷ Donnay, J. D. H., La biréfringence de forme dans la vaterite: *Annales Société géol. Belgique*, 1936 (in press).

with index 1.30, 23 per cent. of a medium with index 1.34. The last figures account for a lowering of the indices to values that depart from the meas-

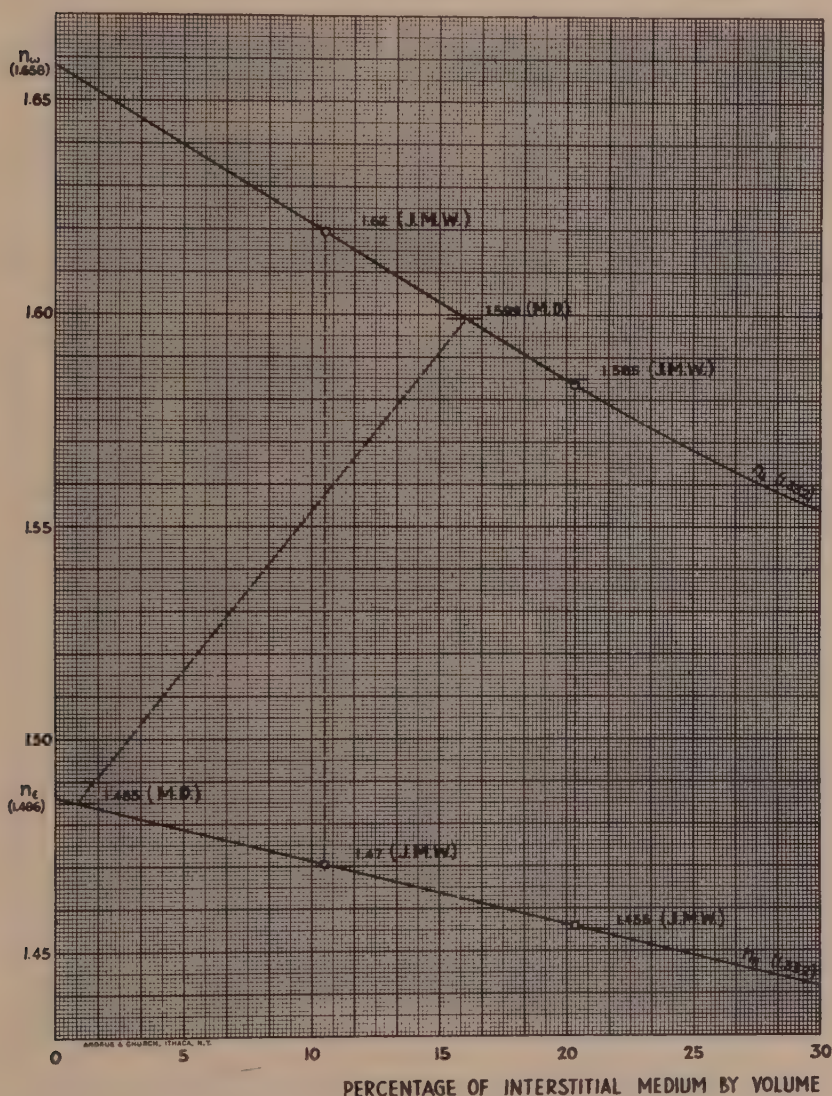


FIG. 13. Action of form birefringence in calcite (=vaterite A) spherulites. Curves showing the lowering of the fiber indices (n_{\perp} and n_{\parallel}) with increasing percentage of interstitial medium (water, index taken as 1.332). Index measurements by Johnston, Merwin, and Williamson (1916) shown by their initials.

ured ones by less than 4 in the third decimal place. Such results are reasonably in accord with what could be expected.

Lowering of indices in spherulitic fibers composed of calcite crystals.—If we repeat the calculations for spherulitic fibers of calcite (= vaterite A) we get the curves shown in Fig. 13. The fibers are supposed to be built up of calcite crystals oriented with their optic axis parallel to the fiber length. The form birefringence is positive.

Johnston, Merwin, and Williamson (1916) give two sets of values for the fiber indices: 1.62 and 1.47 (in the text, page 488, *op. cit.*), 1.585 and 1.455 (in the footnote, same page). They write that the latter were obtained in one case only. For the more usual material, they report (*op. cit.*, p. 489) the presence of about 10 *per cent.* (by volume) of interstitial medium, "mainly water."

The agreement with the theory is perfect. The curves, drawn for an index of the medium equal to 1.332, show that the values which lie on the same vertical between 10 and 11 *per cent.* by volume of the medium depart from the measured values (1.47 and 1.62) by less than 2 in the third decimal place. Again for the values obtained in the isolated case (1.455 and 1.585), theoretical values not off by more than a few units in the third place are seen to lie on the vertical corresponding to 20 to 21 *per cent.* by volume of the medium.

The same figure shows that the fiber indices we have measured on our CaCO_3 spherulites cannot be accounted for if calcium carbonate occurs in the calcite modification.

Lowering of indices in fibers of strontium carbonate spherulites and barium carbonate spherulites.—Similar curves have been plotted for strontium carbonate fibers and for barium carbonate fibers. The results are disappointing. In neither case is it possible to account for the lowering of the indices by means of the above theory.

Strontium carbonate fibers have measured indices of 1.510 and 1.595, while strontianite has the indices 1.520 and 1.667. It is striking fact that the low index, $n_{||}$, is lowered much less than the high index, n_{\perp} . We have tried crystal orientations that would lead to positive or negative form birefringences; we also tried to consider strontianite as the "medium" interstitial between the gelatine as well as the more obvious conception of strontianite crystals separated by the gelatine; in no case can the facts be reconciled with the theory.

The same conclusion holds for barium carbonate fibers. It is reasonable to suppose, then, that some other factor, besides crystal orientation along the fiber and form birefringence, must intervene in the alteration of the values of the indices.

Similar phenomena in fibrous minerals.—In his recent paper on ishkyl-

dite,¹⁸ Syromyatnikov presents interesting speculations on the optical properties of fibrous minerals. He attempts to explain the properties of α -chrysotile and ishkyldite by means of the Wiener theory. Quoting him:

The intermediate medium for this mineral (α -chrysotile) is probably the absorbed water, filling the interstices between the elementary fibers. Its refringence must be lower than that of chrysotile. The integral index of refraction will lie between the refractive index of water and that of chrysotile. *As the fibers of chrysotile are continuous throughout their length, the refractive index N_0 must remain constant and independent of the surrounding medium.* Hence, we shall be interested chiefly in the optic vector across the length of the fiber.

The statement, which we have placed in italics in the above quotation, shows clearly that Syromyatnikov was facing the same difficulty as we are, when trying to explain the fact that the refractive index parallel to the fiber length, n_{\parallel} , is very little lowered by the action of the interstitial medium.

Conclusion.—If Syromyatnikov's statement with respect to the value of n_{\parallel} is correct, it will provide an easy way out of our difficulty. The splinters of our spherulites are also made of fibers continuous throughout their length, and hence n_{\parallel} should not be lowered to any great extent, whereas n_{\perp} would be the lowered value of n_0 resulting from the presence of the interstitial medium.

One last possibility is that the spherulites are in a state of strain, resulting from their mode of preparation in gelatine. One fact of observation (reported in our second paper) is that some spheres break up, when rubbed, with almost explosive violence. This is true of lead sulfate especially, but has also been noticed in spherulites of calcium carbonate. This is taken to indicate that the spherical aggregate is under strain. Now strain will induce additional "accidental birefringence," the refractive indices being altered still further. We have not attempted to give this last factor a quantitative expression, fearing that the mathematical treatment involved would be beyond our competence.

The good agreement found between the theory and the experimental data of Johnston, Merwin, and Williamson, in the case of vaterite *A*, may be explained by the fact that their spherulites, grown in aqueous solutions, were not in a state of strain, and hence no additional accidental birefringence was involved.

SUMMARY

The paper presents the results of further study of the shape and the optical properties of artificial spherulites and related aggregates, grown by diffusion and metathesis in gelatine.

¹⁸ Syromyatnikov, F. V., Ishkyldite, a new structural variety of chrysotile: *Am. Mineral.*, vol. 21, pp. 48-54, 1936.

Although these aggregates may vary widely in external shape, most of them belong to one and the same type of "aggregation," due to radiating crystallization. An hypothesis is advanced as to the mechanism of formation of spherulitic structures, *viz.*, that a continuous branching out of fibers makes an aggregate look like a sheaf "opening" at both ends, in fan-like manner, as growth proceeds until spherical shape is attained or, at least, approximated. The hypothesis rests on the observation of a series of forms interpreted as transitional stages. The central "Zweiblatt," described by Popoff in two-dimensional spherulites, finds its counterpart in our three-dimensional structures (in one case).

The *spherulitic figure* (x-nicols, parallel light) is compared with the interference figure observed (x-nicols, convergent light) in a uniaxial crystal cut at right angle to the optic axis. The circular outline of spherulites, viewed in plane polarized light, appears elliptically distorted when the refractive index of the mounting medium falls between the principal indices of the fibers. Contrary to Spangenberg's idea this optical effect can give no information on the indices of the crystals constituting the fibers.

The optical unit of the structure is the fiber, which behaves as a uniaxial entity. Its refractive indices, however, differ from those of the crystalline species that builds up the fiber. Various factors are considered to explain this discrepancy: crystal orientation along the fiber, form birefringence, and accidental birefringence due to strain.

ERRATA IN PREVIOUS PAPER

Am. Jour. Sci., vol. 23, 1932, page 427, line 6, read $n_\gamma = 1.615$, $n_\alpha = 1.511$, instead of 1.677 and 1.529.

ibid., page 431, signs of anglesite, barite, celestite, should be (+) instead of (-).

IGNEOUS ROCK NAMES AND THEIR EVALUATION

JOHN C. HAFF, *Columbia University, New York City.*

The writer has recently had the privilege of assisting in the revision of a glossary of the nomenclature of petrology. Certain observations were made bearing on the validity, on the mode of establishment, and on the applicability of many rock names in the literature during the course of this work. It is perhaps not generally realized how many varying reasons have been advanced by different authors for proposing a new rock name. An examination of the literature of petrology has shown that the bases of proposal have been greatly diversified. The reasons and arguments set forth, in certain individual but highly typical instances, seem to have been somewhat inadequately integrated with petrologic systems. The more specious names seem to have been advanced without reference to any system or standards whatever. But it may be stated that most authors, at the time of proposal, seem to have been fully aware of their responsibility. Many writers have been extremely diffident about coining a new rock name, but have eventually given way, as the multitude of igneous rock names indicates.

It is regrettable that there are no formal nomenclatorial rules available at present to petrologists, and this is probably a reflection of the stage of development in petrology toward a more rational system. Some salient standards for the naming of rock types have long been needed. But their establishment is obviously the work for a committee, or committees, and will require the concerted efforts of many authorities before completion.

The intention of this paper is, first, to emphasize the great need for precaution in the proposal of additional rock names. It is hardly necessary to call attention to the host of equivocal and loosely defined terms which have appeared by the misuse of prerogative. Second, it is hoped that the observations made here will be of some assistance to the student in evaluating some of the different types of rock names already in the literature.

Locality names appear to be the most abundant, and in many cases the most offensive, of all rock names. This is especially true inasmuch as many of them have obviously been proposed without sufficient consideration of their often limited application. Probably most petrologists agree that the number of usable, fundamental rock names is relatively few. It is well known that the increase, particularly of locality names, and the use of innumerable varietal and sub-varietal names has led to duplication and confusion. It may be said that the promiscuous coining of new names is an offense of the past, or that useless names disappear of their own accord. Actually it appears that some authors have accepted

certain names, and different authors others, and all have used the lot with conspicuously varying meaning. Those names which are quite generally accepted are frequently used with meanings which are deviations from the original sense, or expansions of it, and even greater confusion has resulted from this practice.

Systematic names are here defined as those rock names which have been established with direct reference to some system of petrology. This system is usually fundamentally chemical but almost always includes certain concepts of rock derivation, geologic occurrence and habit. Systematic names as the familiar granite, gabbro, syenite, basanite and trachyte, for example, have been established by general acceptance and recognition of their broad worth and applicability. In most instances these relatively ancient names antedate the development of our modern system or systems of petrology, but have nevertheless been the framework around which the nomenclature has been established. Less widespread rock types and those of restricted or special occurrence have necessarily been referred to, and clustered around, these fundamental names.

In addition to those systematic rock names the origin of which may be said to be related to the earlier days of petrography, there are other systematic names which may be called the petrographic series type. In his monograph on the grorudite-tinguaite series of the Christiania district, Brögger¹ established the existence of a petrographic series of dike rocks composed of closely related members, chemically and mineralogically transitional to one another. The most constant mineralogical characteristic of the members of this series is the occurrence of potash-feldspar, microperthite and potash-soda-feldspar, with abundant aegirine. Soda-amphiboles are also of widespread occurrence and quartz is generally present especially in the rocks of the grorudite, or acid end of the series. Nepheline is a quite constant mineral, although in variable amounts in the rocks within the limits of the series, and subordinate or lacking toward the more acid extremes. Plagioclase is typically absent and biotite usually occurs in only small amounts. The chief chemical characteristic of this series is the predominance of soda and potash, the potash always being less in amount than the soda, and the comparative recession of magnesium and calcium. In the Christiania region this series ranges, in terms of the best developed rock types, from the more acid grorudite through solvsbergite to tinguaitite.

From a study of the ratios of the variations of the potash and soda, from the more acid rocks to the more basic extremes, Brögger was able

¹ Brögger, W. C., *Die Eruptivgesteine des Kristianiagebietes. I. Die Gesteine der Grorudit-Tinguait-Serie: Videnskabselskabet's Skrifter, I. Math.-Naturv. Kl., 1894, No. 4, pp. 159-186.*

to calculate the approximate composition of a hypothetical basic end-member of the series. The calculated end-member was not, however, found by him among the dike rocks of the Christiania region. But a dike rock from Beemerville, New Jersey, earlier described by Kemp² as an "elaecolite-porphry," was found to have a chemical composition nearly like that of the hypothetical end-member. From the description of this rock, Brögger decided that it also constituted a satisfactory mineralogical equivalent of his calculated end-member. He therefore named this rock *sussexite*³ from its occurrence in Sussex County, New Jersey.

This is one outstanding instance in which sufficient chemical data, pertaining to a closely related series of rocks, were available for study. Moreover, chemical gradations between members of this series were satisfactorily smooth and well-defined, so that transitions could be definitely followed. Hence the facts necessary to establish the composition of the projected end-member could be determined. The validity of the calculations was in this instance immediately verified by reference to a description of an actual occurrence of a rock comparable to the desired end-member. Even if no example of an end-member had been known or described at the time of proposal of the new name, a prophetic name might have suitably been given, in consideration of the strong probability of its being discovered. This is an example of what might be called a cultivated rock name.

In 1887, Rosenbusch, having considered the well-established occurrence of the extrusive nepheline-tephrites, was able to postulate the discovery of their plutonic equivalents on grounds which were purely systematic. He therefore felt free to propose the name *theralite*⁴ (Gr. eagerly sought rock) because the weight of probability, systematically arrived at, indicated that these equivalents would eventually be found. Wolff⁵ had, indeed, already described certain nepheline-plagioclase rocks from the Crazy Mountains of Montana, which might have been considered the desired rock type. But the description of Wolff, which indicated that the Montana rocks occurred as dikes and in small bodies of uncertain structural relationship, did not satisfy Rosenbusch that this occurrence was actually plutonic in character. In 1903, Bauer⁶ described a highly mela-

² Kemp, J. F., The elaeolite syenite near Beemerville, Sussex Co., N. J.: *Trans. New York Acad. Sci.*, vol. 11, p. 66, 1891-92.

³ Brögger, W. C., *op. cit.*, p. 173.

⁴ Rosenbusch, Harry, *Mikroskopische Physiographie der Massigen Gesteine*, 2nd Ed., p. 248, 1887.

⁵ Wolff, J. E., Notes on the petrography of the Crazy Mountains and other localities in Montana Territory: *Northern Transcontinental Survey*, R. Pumpelly, Director, 1885.

⁶ Bauer, Franz, Petrographische Untersuchung des Duppauer Theralithvorkommens: *Tschermaks Min. und Pet. Mitt.*, N.F., vol. 22, pp. 266-296, 1903.

nocratic rock from the Duppau region in Bohemia, with a hypidiomorphic-granular structure, composed essentially of titanaugite, labradorite, nepheline, and barkevikite, with some biotite, olivine, and a little orthoclase. This rock, being typically massive and plutonic in habit, was accepted by Rosenbusch as the type theralite. Hence the strong basis of accumulated facts and observations, upon which Rosenbusch had established his petrologic system, had accurately indicated the probability of this plutonic occurrence. Such dependence upon, and integration with, the well-established concepts of a highly refined petrologic system is indispensable to the successful classification of a new rock type.

A series of related rocks has sometimes been given the name of the most representative or widely developed member of the series exposed in the type region. Or a regional series, as contrasted with a petrographic series, has been called by the name given that rock representing a distinctive type to which other rocks of smaller bulk development or of less well-defined mineralogy could best be referred. To attach a regional series significance to a rock name involves a geographical connotation and implies the distribution of the associated rocks within a fairly well-defined area. A stratigraphic significance may also be implied, even in the case of igneous rocks, inasmuch as the members of a series of very closely related igneous rocks may have been, and probably oftentimes were, consolidated within the limits of a single epoch of vulcanism.

The Geological Survey of India has made frequent use of series names of this type for simplicity and convenience in mapping large areas. The charnockite series of Holland is an example. The name charnockite was applied by him to a quartz-feldspar-hypersthene-iron-ore rock of igneous origin, found in Peninsular India. Holland⁷ applied the name charnockite-series to that group of phanerocrystalline igneous rocks in India which gave evidence of being magmatically related in time and space to the distinctive hypersthene rock, charnockite. The related rocks of this series, by definition, may range through to rocks as basic as norite and pyroxenite, but all show distinctive family resemblance to one another. According to Holland this resemblance is usually expressed by the constant occurrence of ilmenite in place of magnetite and by the absence of titanite throughout the series. These chemical and mineralogical resemblances are thought to be the expression of a true consanguineous relationship between the member rocks. In the case of this charnockite-series, as the term was applied by Holland, those related rocks of the series, which are mineralogically similar to rocks frequently developed elsewhere, are ordinarily designated by the names for those aggregates,

⁷ Holland, T. H., The charnockite series, a group of Archean hypersthene rocks in Peninsular India: *Mem. Geol. Surv. India*, vol. 28, part 2, p. 128, 1900

as norite or pyroxenite. The rocks of the series may occur as dikes or plutonic masses, for the application of the qualifying term "series" does not effect a restriction of the term to rocks of any special occurrence. Holland himself preferred to restrict the application of the term charnockite-series to those rocks developed only in India, unless identical relationships could be proved for rocks elsewhere. He states:⁸

"But unless a similar formation found in another country, can be proved to be a genetic relation of the typical exposures described in this paper, it is hoped that the name charnockite will never be used outside India. And; Charnockite is a convenient name for a quartz-felspar-hypersthene-iron-ore-rock in the charnockite series, and not a name for any hypersthene-granite occurring in other petrographical provinces."

In a similar manner, Fermor⁹ applied the term kodurite-series to a group of closely related manganiferous rocks developed in the Madras Presidency, India. The typical rock of this series is coarsely crystalline, composed of potash-feldspar, manganese garnet and apatite. It may vary in texture from granitoid to pegmatitic. The potash-feldspar content of the original rock had to be calculated originally, as all the feldspar appeared to have been replaced by opal in the type material. The mineralogy of kodurite was so distinctive that varietal names, as quartz-kodurite and pyroxene-kodurite, were used by Fermor for rocks varying from the type because of the occurrence of these minerals. The origin of the rocks of the kodurite series is not definitely established, but in the usage of the Indian Survey this does not invalidate the application of the series terminology. As there used, the term series is applied to both igneous and metamorphic rocks if the genetically distinctive features of the related rocks can be established.

Group names are those used to link together for convenience a number of rock types characterized by a particular mineral or mineral combination. A group name also implies a genetic similarity of structure among the rocks included in any given group. In the use of group names relatively little latitude in the mineralogy of the rocks included in a group is permissible, as compared with the rocks of a regional series such as just described. Those rocks assigned to a particular group need be in no way related, however, other than by the fact of their mineralogical traits. No geographic significance is attached to the rocks placed within a mineralogically defined group, for they are allocated to that group irrespective of their distribution or space relationship. Although artificially arrived at, typical group names may be of convenience in classification. The perknite group of Turner¹⁰ was defined as including those massive,

⁸ Holland, T. H., *op. cit.*, p. 131.

⁹ Fermor, L. L., *Rec. Geol. Surv. India*, vol. 35, part 1, p. 22, 1907.

¹⁰ Turner, H. W., Perknite (Lime-Magnesia Rocks): *Jour. Geology*, vol. 9, p. 507, 1901.

coarsely crystalline and relatively basic rocks characterized by the association of amphiboles and monoclinic pyroxenes. He included under this term rocks bearing rhombic pyroxene, olivine, and feldspar, in small amount, with the usual accessories as biotite and iron-ores. According to Turner, who permitted considerable freedom in the application of this term, only one of the specified essential minerals need be present in the rocks, this expansion allowing the inclusion of nearly monomineralic rocks as well. Such rocks as pyroxenite, hornblendite, websterite, diallagite, and bahiaite, would therefore be included in the perknite group of lime-magnesium rocks.

Certain terms have been given wide application when introduced, some being originally so broad as to possess practically group or series standing. Certain nepheline bearing rocks from Brazil, of widely ranging lithology, were called by Derby¹¹ jacupirangite. Under this term he included rocks composed of magnetite alone, i.e., ores, rocks composed of magnetite and accessory pyroxene, pyroxene rocks with accessory magnetite, and pyroxene-nepheline rocks. All these types may contain biotite and olivine as accessories. In the Brazilian occurrence the rocks were all part of the same mass and transitional to one another, and might indeed have been called a series. The range of these rocks included under the same name should be noted. In fact the mineralogy of any type rock was not stipulated by Derby, the intention being apparently to use the name with the very broadest significance. As used at present, though, the term jacupirangite may be said to be restricted to a nepheline-titanaukite-ilmenite rock. This restriction has been applied chiefly in recognition of the unusually high and characteristic titanium content of the rock, and the abundance of nepheline. Derby had hinted at this himself, and stated that the most constantly ranging mineral was a titaniferous pyroxene, but he apparently was not aware of the composition of his so-called magnetite, now believed to have been ilmenite. This is an instance in which petrologists have been quick to discern the more critical features of a suite. Hence they have proscribed and limited the term jacupirangite giving it more critical significance.

Many older rock names of formerly variable application have never been re-defined, but have acquired approval and specific meaning through usage and general concurrence. Other rock names have undergone formal re-definition, in some cases repeatedly. Many of the older rock names as originally proposed were applied to rocks inadequately described for more modern purposes. Re-definition of some terms has given them addi-

¹¹ Derby, O. A., On the magnetite ore districts of Jacupiranga and Ipanema, Sao Paulo, Brazil: *Am. Jour. Sci.*, 3d ser., vol. 41, p. 314, 1891.

tional significance by establishing the distinguishing features of the rocks with greater clarity in the light of more recent opinion. In cases where a rock name has had very loose and perhaps contradictory application, re-definition has been imperative if the term were to have any value. In such instances the re-definition may, it appears, quite excusably violate the intention of the author of the name. De Lapparent¹² coined the name monzonite in 1864, and it was for a long time used as a collective name for different rocks of the Adamello region. It was also used as a series name, and applied by Rosenbusch to a particular group of augite syenites. No unanimity of application ensued for many years, during which time the name monzonite was used for hornblende-plagioclase rocks, for diorites, augite diorites, and augite syenites, i.e., for rocks which were either plagioclase or orthoclase bearing. Ultimately Brögger re-defined the term in 1895, establishing monzonite as a transition rock between the syenites and the diorites, and as one containing typically both plagioclase and alkali-feldspar. To quote:

"Die Monzonite charakterisiren sich eben dadurch, dass sie weder zu den Orthoklas-Gesteinen noch zu den Plagioklas-Gesteinen, sondern zu einer Übergangsgruppe oder Zwischengruppe zwischen beiden gehören, sie sind eben: Orthoklas-Plagioklas-Gesteine.¹³ . . . Es ist mit anderen Worten nach meiner Ansicht nothwendig, zwischen den Orthoklasgesteinen und den Plagioklasgesteine, oder wie es jetzt correcter heissen muss: zwischen den Alkalifeldspath-Gesteinen und den Kalknatronfeldspath-Gesteinen, eine Übergangsordnung von Alkalifeldspath-Kalknatronfeldspath-Gesteinen einzuschieben."¹⁴

This term, as re-defined by Brögger, appears to embody the essential features of the more recent applications of the term.

Occasionally some rocks, as originally described, were stated to contain minerals later found to have been misidentified. Sometimes a name has been proposed for a rock, the mineralogy of which was substantially correct as given, though the interpretation of its origin has proven incorrect. Re-definition does not, apparently, proceed by any established rule but at the complete discretion of the writer. Names applied originally to rocks of a definite mineralogy have been re-defined so that a structural signification is given the term. Blum¹⁵ proposed the name foyaite in 1861 for a syenite-like rock from Mount Foya, Portugal, distinguished by the presence of abundant nepheline. This typically granitoid rock composed of orthoclase, nepheline, and hornblende (later dis-

¹² de Lapparent, J., *Mémoire sur la constitution géologique du Tyrol méridional: Annales des Mines*, vol. 6, p. 259, 1864.

¹³ Brögger, W. C., *Die Eruptivgesteine des Kristianiagebietes. II. Die Eruptionsfolge der Triadischen Eruptivgesteine bei Predazzo in Südtirol: Videnskabselskabs Skrifter, I. Math.-Naturv. Kl., 1895*, No. 7, p. 21.

¹⁴ *Idem.*, pp. 22-23.

¹⁵ Blum, R., Foyaite, ein neues Gestein aus Süd-Portugal: *Neues Jahrb.*, p. 426, 1861.

covered to be aegirine in part), might have been called simply nepheline syenite, though a new term was applied. In 1890 Brögger¹⁶ called all those nepheline syenites distinguished by trachytoid structure, foyaïtes. He took no account whatever of their composition, which thus might range through all the varietal types within the nepheline syenite group. Accordingly, the name foyaïte, in the sense of Brögger, no longer signified the type rock. At the same time he applied the term ditroite to those nepheline syenites with typically hypidiomorphic-granular structure; this change also constituted a re-definition, since Zirkel¹⁷ defined ditroite as a sodalite-bearing nepheline syenite. Hence in both instances there is the connotation of a structural characteristic attached to a rock name, which was previously applied to a certain mineralogical aggregate.

Perhaps the original occurrence of a rock has not afforded enough exposures, or sufficiently varied material, to enable the author of a name to make a comprehensive study. Perhaps then, a later writer has been able to establish the relationships of the rock more advantageously. This was done in the case of arkite, a holocrystalline porphyritic leucite (pseudo-leucite)-nepheline-pyroxene-garnet rock from Arkansas, first mentioned by Williams,¹⁸ later analyzed, re-described, and named by Washington.¹⁹ The guiding principal in re-definition seems to be an elaboration of the mineralogy, structure, origin, and relationships of a rock to establish more clearly its systematic place.

In certain cases one may resort to provisional names. The authors of names of this type have recognized that their description, presented at the time of proposal of the name, was inadequate to establish fully the distinguishing characteristics of the rock. Rocks of limited distribution, of confused relations, of dubious affinity or extreme peculiarity, may thus be given names stipulated at the outset to be tentative. Accordingly the reader is admonished not to apply the name indiscriminately and is expected to refrain from its rigorous application until the relationships of the rock can be set forth with greater clearness pending additional study. Harker,²⁰ for example, described as mugearite an unusual basalt bearing chiefly oligoclase, with some olivine and lesser augite. It may be assumed that he did not wish to commit this name to

¹⁶ Brögger, W. C., Die Mineralien der Syenitpegmatitgänge der Südnorwegischen Augit- und Nephelinsyenite: *Zeit. Krist.*, vol. 16, p. 39, 1890.

¹⁷ Zirkel, Ferdinand, *Lehrbuch der Petrographie*, p. 595, 1866.

¹⁸ Williams, J. F., The igneous rocks of Arkansas: *Ann. Rept. Geol. Surv. Arkansas*, 1890, vol. II, pp. 274-276.

¹⁹ Washington, H. S., The foyaïte-ijolite series of Magnet Cove; a chemical study in differentiation: *Jour. Geology*, vol. 9, p. 617, 1901.

²⁰ Harker, Alfred, The Tertiary igneous rocks of Skye: *Mem. Geol. Surv. Great Britain*, p. 257, 1904.

circulation until further research had been completed, and hence applied a name specified to be provisional.

Terms of this type have occasionally proved too tempting to some writers, namely those prone to make rather hasty comparisons. Hence some provisional names may often be very casually applied despite warnings that the restricting genetic features of the rock are not yet sufficiently plain. Certain delicate questions of priority are sometimes involved in the proposal of provisional names. But the use of such tentative names, on the whole, seems to be in accord with more meticulous and deliberate scientific procedure.

Somewhat in the same category as provisional names are those rock names which are stipulated to be of only local application. Tyrrell²¹ has called a hyalocrystalline anorthite-enstatite-augite andesite dike rock from the Clyde area, Scotland, *cumbraite*. He seems to have felt that this locality name would be of convenience in describing certain rocks of this particular district. But he preferred that the name should not be freely used until the more widespread distribution of actual *cumbraite* could be proved in other petrographic provinces and periods. Hyaline rocks of analogous composition to *cumbraite*, but associated as facies of larger intrusives, would of course be given names suggestive of their relationship to the parent mass. The proposal of a new term for hyaline rocks is, in the usage of Tyrrell, permissible only when they are of individual mass development.

The foregoing discussion has concerned certain types of igneous rock names proposed or re-defined in accordance with generally satisfactory procedures. Examples are given below of additional types which, it seems to the writer, were proposed on more insecure grounds. Attention will be directed especially to the difficulties arising from the proposal of certain names with the use of what are believed to be rather questionable principles.

The term "porphyry" has had wide application in mining usage, having been applied in general to an intrusive rock as distinguished from an extrusive one. It has thus been easy to attach this qualifying term to names like rhyolite and andesite, and to compound names signifying their intrusive equivalents. As a result there has crept into the literature many free terms which some authors do not apply unless the occurrence of the rock is known to them. Hence certain rocks might be distinguished as rhyolite porphyries, granite porphyries or quartz porphyries, if the intrusive occurrence is known, although laboratory study might not divulge evidence establishing the type of occurrence independent of in-

²¹ Tyrrell, G. W., Some Tertiary dykes of the Clyde area: *Geol. Mag.*, 6th. dec., vol. 4, p. 313, 1917.

formation other than that derived from thin sections. This represents a conflict between a so-called practical application and a generic application in greater accordance with systematic usage. The practice of giving a separate name to equivalent hypabyssal types has entered into many schemes of classification, because on a petrographic basis and with supporting chemical data, it is sometimes possible to discern features peculiar to hypabyssal rocks. Nevertheless, an anomaly arises, for, as in the case of terms like andesite and andesite porphyry, the porphyritic structure may be understood in the first instance, while it has seemed incumbent on some to express the structure in the case of the second term.

To make matters worse, such terms as quartz porphyry and granite porphyry are used with no uniformity even by accomplished petrologists. Johannsen²² refuses to use the term quartz porphyry for acid dike rocks, because it has so long been used by European geologists for pre-Tertiary effusive equivalents of the granites. He groups rocks like the granophyres under the granite porphyries. On the other hand, Harker²³ has made the granite porphyries a sub-type under his major group of acid hypabyssal rocks, the quartz porphyries. He applies this latter term with a broad significance and considers granite porphyries, granophyres, and acid pitchstones varieties which can best be grouped with the quartz porphyries.

Gradations in a dike feeder to a flow, or a dike apophysis from a plutonic body, are admittedly difficult to follow and delimit. A dike rock, at a level at which it has obtained typical development may possess fairly distinctive internal structures. The ocellar structure common to certain lamprophyres may be taken as an example. However, the structures of dike feeders and their correlated flows must grade imperceptibly into structures typical of flows in the extravasated rock, and into more massive structures in those parts of the dike in closer proximity to the parent plutonic mass. Another variable, in the form of increasing differentiation with increasing distance from the parent mass, may be assumed to influence the composition and mineralogy of the rock. It is here that one must consider the hypabyssal concept as set forth by Brögger.²⁴ He used this term, in contradistinction to the term "Ganggesteine" of Rosenbusch, as signifying those rocks which are structurally

²² Johannsen, Albert, *A descriptive petrography of the igneous rocks*, vol. II, Quartz-bearing rocks, p. 286. Chicago, 1932.

²³ Harker, Alfred, *Petrology for students*, 6th Ed., p. 102, 1923.

²⁴ Brögger, W. C., Die Eruptivgesteine des Kristianiagebietes. I. Die Gesteine der Grorudit-Tinguait-Serie: *Videnskabselskabet's Skrifter*, I. Math.-Naturv. Kl., 1894, No. 4, pp. 123-124.

and mineralogically comparable, irrespective of their development as dikes, as marginal facies of larger intrusives, or as flows. Inasmuch as certain rocks petrographically indistinguishable from one another had been found by him in these varying environments, he considered "hypabyssal" a more accurate and inclusive term.

As the result of the application, in certain petrographic systems, of different names to nearly equivalent rocks of varying occurrence, writers have sometimes proposed new names for rocks structurally and mineralogically comparable, though consolidated under somewhat different conditions. The fact of a differing geologic environment alone appears to have been considered a reason for the new name. At times differences of structure and mineralogy between the rock described and comparable rocks of different occurrence have been most obscure. Pirsson²⁵ investigated a rock fragment, from a core boring put down on the island of Bermuda, which he believed to be derived from a flow. This was of a moderately porphyritic rock composed of biotite phenocrysts in a nearly isotropic groundmass consisting of analcite, sanadine and nepheline. With this effusive biotite-rich rock there were associated types containing considerable augite. The biotite-bearing lava was lamphrophyric in character and compared most closely with ouachitite, although the latter rock had been up to that time known only in dikes. Pirsson was most diffident about proposing a new name merely on the basis of occurrence but conceded the point thus:

"... and those who demand a separate name for the effusive rocks as contrasted with the intrusive ones, would object to calling it ouachitite, which it is most like in composition and in genesis. The name of bermudite is suggested for consideration, to obviate this difficulty."²⁶

It is difficult to reconcile such usage as this, although it has often had the approval of systematists. But it is fitting to point out the absence of authoritative agreement on the validity of the concept of the consolidation of discrete dike types as distinguished from extrusive types. Such a concept is opposed by the occurrence of hypabyssal facies and the gradual structural variations between an intrusive and its related extrusive body. From a petrographic point of view, in the absence of some significant distinguishing structure, mineralogy, or chemical composition, a new name should hardly be proposed for a related rock merely on account of a difference in occurrence.

Peculiarity of occurrence alone has been used as a basis for the proposal of certain rock names. C. W. Knight described a crystal-tuff, occurring in Alberta, composed of fragments of analcite and potash

²⁵ Pirsson, L. V., *Geology of Bermuda island; petrology of the lavas: Am. Jour. Sci.*, 4th ser., vol. 38, pp. 331-344, 1914.

²⁶ *Idem*, p. 340.

feldspar. The mineralogical and chemical composition of this rock was found to approximate that of an analcite-trachyte. For such a tuff Knight proposed the name blairmorite.²⁷ Thus a derivative of a well established rock type, analcite-trachyte, has been given a separate name in no way suggestive of the composition of the derivative rock, of the rock from which it was derived, or of the geologic occurrence. One small rock fragment of analcite-trachyte was found, but no special name was proposed for it at that time. To the writer it seems ill-advised to create a new name on the rather insecure basis of the pyroclastic occurrence of a rock composed of fragments of an original rock of known systematic place. According to this principle any pyroclastic rock approximating any known rock in bulk composition would be deserving of a new name.

Washington and Larsen used the name arapahite for a fragment of magnetite-basalt. They were in part influenced by considerations of the unusual occurrence of the rock. Outcrops at the type locality appear to have been very poor, but it was thought that the main body of the rock containing their specimen was a breccia. They state:

"While the rock would logically be called a magnetite-basalt, on account of its extrusive character and mode, yet it is so unique as to occurrence and composition, that a special name seems to be justified. For this that of arapahite is suggested."²⁸

The writer has no quarrel with establishing this name on a chemical basis, but the validity of using the peculiarity of occurrence as one criterion for proposal seems open to question, especially as the true nature of the form of occurrence was at that time doubtful.

A rock from New Zealand has been called wilsonite and the name ascribed to Henderson.²⁹ According to a description by Sollas it contained chiefly crystal fragments of acid plagioclase, pyroxene, biotite and quartz, with lithic fragments showing spherulitic growths and others probably of andesite. These were contained in a mottled black and white matrix of glass fragments, some of which had perlitic structure. All of these crystal and lithic elements were cemented by an isotropic glassy base. Opinion differed as to the origin of this rock. Henderson, however, found some fragments of fairly fresh andesite and largely on this basis believed it to be a tuff. Other investigators had considered it a brecciated flow rhyolite. Irrespective of the merits of either case it seems indiscreet

²⁷ Knight, C. W., Analcite-trachyte tuffs and breccias from southwest Alberta, Canada: *Canadian Rec. Sci.*, vol. 9, p. 266, 1904 (1905).

²⁸ Washington, H. S., and Larsen, E. S., Magnetite basalt from North Park, Colorado: *Jour. Washington Acad. Sci.*, vol. 3, p. 452, 1913.

²⁹ Henderson, J., The geology of the Aroha subdivision, Hauraki, Auckland: *New Zealand Geol. Surv., Bull.* 16, New Ser., p. 70, 1913.

to permit the circulation of a new term for a rock admittedly of disputed origin. It has been argued that convenience in discussion of controversial types demands the use of a short term and hence constitutes a basis for using a new name. But if the term is used, the writer feels that it should be made quite clear that it is of the most tentative sort. There are instances in which a new term was proposed although at the time it was impossible to give a correct interpretation of the rock due to lack of knowledge or misidentification of minerals. But at least such terms were made in oblivion of the actual relations, circumstance rendering it impossible to obtain the correct data.

Varietal names have generally been proposed because the rocks under consideration were thought to present a mineralogical or structural departure, in some arbitrary amount, from the type to which the rock given the new name was referred. Evidently some authors have believed that the variant rock warranted a new name solely because of these differences, and often little effort has been made to specify the significance and relationships of the varietal features. Some examples of these varietal names are included here and contrasted with their mineralogically qualified equivalents.

<i>Type</i>	<i>Variety</i>	<i>Varietal Name</i>
Ghibelite	biotite-ghibelite	ponzite
Monzonite	olivine-monzonite	kentallenite
Hypersthene	hornblende-hypersthene	bahiaite
Basalt	magnetite-basalt	arapahite
Bostonite	quartz-bostonite	lindoite
Nepheline-syenite	albite-rich	canadite
Peridotite	enstatite-peridotite	saxonite
Essexite	olivine-rich essexite	montrealite
Analcite-basalt	biotite-rich	ghizite
Limburgite	leucite-limburgite	batukite
Nepheline-syenite	feldspar-free	monmouthite
Soda-granite	hypersthene-rich	birkremite
Diabase	albitic	holyokeite
Theralite	feldspathic	covite
Picrite	pyroxene-rich	ankaramite
Theralite	leucocratic	rouvillite
Phonolite	microperthitic	apachite

Some varietal names based on structure.

<i>Type</i>	<i>Variety</i>	<i>Varietal Name</i>
Alaskite	porphyritic	tordrillite
Hornblendite	poikilitic	cortlandite
Nepheline-syenite	trachytoid	foyaite
Nepheline-syenite	hypidiomorphic	ditroite

As a rule it has been construed that the varietal rock constituted a type, which, being transitional to a better established type, was therefore an intermediate rock, and hence that a new term was a refinement of the nomenclature. If this practice were carried to the extreme, an infinite number of rock names might result, each representing a deviation of variable, arbitrary amount (and often of the most subtle significance), in any direction whatever from the type.

Complaint has sometimes been made of the cumbersomeness and lack of euphony of compounded names beset with qualifying mineral names. But in many cases a greater sacrifice of clarity and lack of understanding has resulted from the multiplication of varietal names. Those varietal names of elusive, inadequate definition, which are expressed in terms of a locality are perhaps the most offensive of all. It is, of course, most difficult to state how much deviation from a type constitutes a basis for the proposal of a new name since so many standards could be applied. The entirely subjective considerations of the author of the new name may sometimes appear most inconsequential to other workers. At the very least it seems that varietal names should be meticulously defined, that pains should be taken to specify what the significant variations are, and the amount of variation should be quantitatively expressed if possible.

Washington³⁰ proposed the name bahiaite for hornblende-hypersthene. The type hornblende-hypersthene had previously been described by Merrill³¹ from Montana. The bahiaite of Washington was stated by him to be closely comparable in mineralogy and structure to Merrill's type material. Merrill believed the term hornblende-hypersthene cumbersome, but considered it preferable to a locality name inasmuch as the rock was the only example of that type known to him. Washington stated, on the basis of this one additional occurrence of the rock in Brazil, that the widespread occurrence of hornblende-hypersthene was thus established. Hence he proposed the locality name bahiaite without any reference to the locality of the type rock described previously by Merrill. This procedure is the opposite of the usual one in which local rock peculiarities and restricted distribution are often emphasized by the application of a locality name. To the writer it seems anomalous to apply a special locality name to a rock on the basis of its widespread occurrence, especially when a properly qualified rock name of known type would suffice. Unquestionably certain locality names are

³⁰ Washington, H. S., An occurrence of pyroxenite and hornblende in Bahia, Brazil: *Am. Jour. Sci.*, 4th ser., vol. **38**, p. 86, 1914.

³¹ Merrill, G. P., Notes on some eruptive rocks from Gallatin, Jefferson, and Madison Counties, Montana: *Proc. U. S. Nat. Mus.*, vol. **17**, pp. 656-659, 1894.

useful in briefly typifying some very distinctive rock types. But the propriety of applying a particular locality name to an admittedly abundant and widely distributed rock seems highly dubious indeed.

The writer feels that it is now beyond the capability of the individual to reconcile the confused usages just indicated with better usage, but suggests that comparisons be made with those more substantial types of rock names earlier mentioned in this paper. Practical application, precedent, individual preference, and varying concepts of rock derivation are all involved in the proposal of a rock name. The evident lack of standards for the proposal of names might be in part circumvented by applying a quantitative mineralogical scheme, necessarily highly arbitrary but desirable petrographically. Ultimately magmas as well as mineral aggregates must be taken into account, inasmuch as those mineral aggregates produced from a given magma may vary with differing environments. A really successful nomenclature will probably be binomial in character and will thereby bring together the petrographic and magmatic attributes of rocks.

CRYSTALLOGRAPHIC DATA, UNIT CELL AND SPACE GROUP FOR BERTHIERITE (FeSb_2S_4)

M. J. BUERGER, *Massachusetts Institute of Technology,
Cambridge, Massachusetts.*

ABSTRACT

Crystallographic data are lacking for berthierite because its imperfect morphological development prevents optical goniometric study. These data, however, have been easily obtained by Weissenberg methods. An equi-inclination study leads to the following results:

Centrosymmetrical symmetry: $mmm = V_h$

Crystal system: orthorhombic

Unit cell:

absolute	ratio
$a = 11.44 \text{ \AA}$	0.810
$b = 14.12$	1.000
$c = 3.76$	0.266 ₃

$Z = 4$ formula weights per cell.

Diffraction symbol: $mmmPna-$.

Space group: Pna (C_{2v}^9) or $Pnam$ (V_h^{16}).

The general geometry allows for 58 possible structures, but a simple intensity relation eliminates all but one, and definitely fixes the space group as $Pnam$. All atoms are on reflection planes ($4c$) with different parameters. Although an intensity-parameter study has not yet been made, a structure has been suggested based upon a substituted sphalerite type framework.

INTRODUCTION

Not only is the crystal structure of berthierite unknown, but crystallographic information is also lacking for this species because of its imperfect morphological development. The mineral occurs characteristically as small needles of irregular diamond-shaped cross-section and without faceted terminations. The needles are minutely striated parallel to their lengths. A crystal, therefore, yields only a continuous band of reflections and spectral effects in the needle axis zone when examined by optical goniometric methods. Crystallographic information regarding berthierite may, however, be easily obtained with the aid of the new Weissenberg x-ray methods.^{1,2,3}

Excellent coördinated density and analytical determinations have been made for the Carpathian Kisbánya berthierite by Zsivny and

¹ Buerger, M. J., The Weissenberg reciprocal lattice projection and the technique of interpreting Weissenberg photographs: *Zeit. Krist.*, vol. **88**, pp. 356–380, 1934.

² Buerger, M. J., The application of plane groups to the interpretation of Weissenberg photographs: *Zeit. Krist.*, vol. **91**, pp. 255–289, 1935.

³ Buerger, M. J., An apparatus for conveniently taking equi-inclination Weissenberg photographs: *Zeit. Krist.*, vol. **94**, pp. 87–99, 1936.

Zombory,⁴ who have kindly made some of their original material available to the writer for the present investigation. The pertinent data for this berthierite are presented in Table 1. The analysis evidently indicates a slight excess of iron over antimony for the ideal formula FeSb_2S_4 .

GENERAL CELL CHARACTERISTICS

Method.—Crystals such as berthierite, whose optical reflections are confined to a confused band in the needle axis zone, may be accurately oriented by optical goniometric means for rotation about the needle axis. The reciprocal lattice can then be completely determined with the aid of a rotation photograph about this axis, together with the Weissenberg resolutions of several adjacent levels. Appropriate crystallographic axes may be chosen according to the symmetries, patterns and dimensions of these reciprocal lattice projections.²

To obtain further data of crystal structural importance, it is necessary to rotate the crystal about one (in the case of a monoclinic crystal) or both of the two crystallographic axes so selected. For space group determinative purposes, only axes in a plane at right angles to the needle axis need be investigated. If such an axis exists (i.e., if the crystal is not triclinic), it can be located by either of two methods. In either case, the crystal is remounted with the needle axis at right angles to the rotation axis, which keeps this perpendicular plane in the rotation axis; the needle axis is also placed parallel with one of the adjusting arc axes of the adjusting crystal holder.³ This allows the crystal to be rolled about its long axis without disturbing its normality with the rotation axis. The amount of rolling required to rotate a crystallographic axis lying in the normal plane, into parallelism with the rotation axis, is controlled in either of two ways:

(a) A very general method may be used if the band of optical reflections and spectra given by the needle axis zone contains peculiarities which can be mapped or otherwise recognized again after the mounting is changed. In this case, a careful map of such peculiarities is prepared before the mounting is changed, using the optical goniometer rotation angles as coördinates. The relation between the optical goniometer zero rotation setting and the Weissenberg rotation zero setting is assumed to be known. It is then possible to refer the map of optical reflection peculiarities to the ω coördinates of the Weissenberg photographs, and therefore to the crystallographic axes which appear on the Weissenberg, needle axis, zero-layer, film. This enables the crystallographic axes to be tied in to optical peculiarities. After remounting as indicated above,

⁴ Zsivny, Victor and Zombory, László, Berthierite from Kisbánya, Carpathians: *Mineral. Mag.*, vol. 23, pp. 566–568, 1934.

the optical reflections are again studied. When the peculiarities have been identified, the band map locates the crystallographic axes in the new orientation and the arc may be adjusted until a crystallographic axis is parallel with the rotation axis.

(b) If, for any reason, it is inconvenient to proceed as above, the new needle orientation may be corrected so that a crystallographic axis is parallel with a rotation axis, by the following method: The crystal holder is placed in position on the Weissenberg or other similar apparatus with the needle approximately normal with the x -ray beam. Laue photographs are then made for a sequence of settings of the crystal holder adjusting arc. These exposures ordinarily need not exceed fifteen minutes and may be made on tiny dental films held in a special holder just behind the crystal. If the crystallographic axis sought is in a symmetry axis or symmetry plane of the crystal (multiplied, if necessary, by a symmetry center, in the case of non-centrosymmetrical crystals), the Laue photograph will show the position pattern which is the cross-sectional projection of this symmetry,² as the correct arc setting is attained. By this means, the crystallographic axis (except in the case of triclinic crystals whose projected symmetry is nil) may be located to within one or two degrees. The accuracy may be still further refined by taking several short rotation photographs in the correct region with a series of settings varying by about half a degree. The correct setting has been attained when the reflections of the several planes of a form record as the same spot on the rotation film.

Symmetry.—For determining the general cell characteristics, use was made of cobalt radiation, to which iron minerals offer little absorption.

The needle axis in berthierite has been designated as the c -axis. Rotation photographs and zero-layer Weissenberg photographs were prepared for rotations about all three crystallographic axes. In addition, first and second layer photographs were made for rotations about both the c - and b -axes.

All Weissenberg photographs agree in displaying the level symmetry C_{21} . This proves that the crystal system of berthierite is orthorhombic, but does not distinguish between the crystal classes mmm , $2mm$ and 222 . (Some intensity data to be introduced later prove, however, that the crystal class is holohedral, mmm .)

Space Lattice Type.—Each of the n -level equi-inclination Weissenberg photographs displays a rectangular level pattern, and the level stacking sequence² is 9 (coincident rectangles). The space lattice type is consequently simple orthorhombic.

Unit cell.—The n -level equi-inclination patterns lead to a set of cell dimensions (checked by the three axial rotation photographs, and re-

fined by ξ_w measurements of high-order pinacoid reflections appearing on zero-level Weissenberg photographs) as follows:

absolute	ratio
$a = 11.44\text{\AA}$	0.810
$b = 14.12$	1.000
$c = 3.76$	0.266 ₈
<hr/>	
$V = 607\text{\AA}^3$	

In the introduction it was noted that the cross-section of a berthierite needle is rudely diamond shaped. It is important to point out that the long diagonal of this diamond is in the direction of the a -axis and not the b -axis as might be expected.

The cell contents may be calculated with the aid of the relation:

$$\text{measured density} = \frac{\text{cell mass}}{\text{cell volume}}$$

$$d = \frac{Z \times f \times 1.649 \times 10^{-24}}{V \times 10^{-24}}$$

where d = measured density

Z = number of formula weights per unit cell

f = chemical formula weight

V = cell volume in cubic Ångströms

Substitution of the appropriate values of d , f , and V for berthierite in this relation leads to a value of Z close to 4 formula weights per unit cell.

Space group.—Comparison of reciprocal cell translations² on n level and zero level equi-inclination Weissenberg photographs shows that in the (100) plane both b and c reciprocal translations are doubled, and that in the (010) plane the a translation is doubled. These multiple translations indicate a glide plane with glide components $b/2 + c/2$ normal to a , and a glide plane of glide component $a/2$ normal to b . The sum total of information obtainable from diffraction patterns may be expressed by the diffraction effect formula² $mmmPna-$. The space group is therefore either $Pna- \approx Pna$ Mauguin (C_{2v}^9) or $Pnam$ (V_h^{16}). It will be shown beyond that certain simplicities in the z coördinates of the atoms are indicated by the spectra from (001) which require space group $Pnam$.

THE CRYSTAL STRUCTURE

Equipoints.—The space group must accomodate 4 formula weights of FeSb_2S_4 per unit cell. The 4 Fe, 8 Sb and 16 S can only be distributed individually among the several equipoints of the two possible space groups, Pna and $Pnam$, as shown in Table 2. The permissible combinations of these equipoints is given in Table 3; their number is very large.

Fortunately, the extremely simple intensity series for the reflections from (001) permits a selection of the correct equipoint combination from the 58 possibilities available. These reflections appear only in even orders and their intensities form a regularly declining series. That the series is one of "regular decline" and not just some kind of gradual decline is attested by the fact that each of its reflections is the most intense on the film for its own $\sin\theta$ region; i.e., the atoms are all in phase for these reflections and are confined to identically populated (001) sheets. The appearance of the spectra only in even orders indicates that these sheets are spaced at $c/2$ intervals. Referred to an appropriate origin, then, each atom must have a z coordinate of $\pm\frac{1}{4}$. This exact coordinate can only be attained by the occupation of a special position which is without z parameters. This eliminates *Pna* immediately, for this space group has no special positions. The only equipoint combination capable of giving all atoms a z coordinate of $\frac{1}{4}$ is combination 47, Table 3. The berthierite structure thus has all atoms occupying the equipoint $4c$ of space group *Pnam*, i.e., all atoms are on the reflection plane with different parameters.

It should be observed that this intensity discussion incidentally proves the holohedral nature of the symmetry of berthierite.

Structure.—The structure of berthierite involves 14 parameters which must be determined 7 at a time. A systematic study of parameter-intensity relations has not, as yet, been carried out. A tentative structure based upon certain reasonable assumptions has, however, been suggested.⁵

TABLE 1

Original analytical results		Results after deduction of insoluble and reduction to 100%	Atomic ratios
S	29.46%	29.78	4.000
Sb	56.06	56.65	2.009
Fe	13.43	13.57	1.049
Mn	trace	—	—
Insoluble	0.33	—	—
99.28		100.00	

$$d_{4^{20}} = 4.65_2.$$

⁵ Buerger, M. J., The crystal structure of berthierite, *Am. Mineral.*, vol. 21, pp. 205–206, 1936.

TABLE 2. EQUIPOINT POSSIBILITIES FOR ATOMS IN BERTHIERITE

Space Group	4 Fe	8 Sb	16 S
<i>Pna</i>	4a	4a+4a	4a+4a+4a+4a
<i>Pnam</i>	4a 4b 4c	4a+4b 4a+4c 4b+4c 4c+4c 8d	4a+4b+4c+4c 4a+4b+8d 4a+4c+4c+4c 4a+4c+8d 4b+4c+4c+4c 4b+4c+8d 4c+4c+4c+4c 4c+4c+8d 8d+8d

TABLE 3. EQUIPOINT COMBINATION POSSIBILITIES FOR ATOMS IN BERTHIERITE

Combination designation	Space Group	4 Fe	8 Sb	16 S	Number of Parameters x y z
1	<i>Pna</i> (C_{2v}^9)	4a	4a+4a	4a+4a+4a+4a	7 7 7
2	<i>Pnam</i> (V_A^{16})	4a	4b+4c	4c+4c+4c+4c	5 5 —
3		"	"	4c+4c+8d	4 4 1
4		"	"	8d+8d	3 3 2
5		"	4c+4c	4b+4c+4c+4c	5 5 —
6		"	"	4b+4c+8d	4 4 1
7		"	"	4c+4c+4c+4c	6 6 —
8		"	"	4c+4c+8d	5 5 1
9		"	"	8d+8d	4 4 2
10		"	8d	4b+4c+4c+4c	4 4 1
11		"	"	4b+4c+8d	3 3 2
12		"	"	4c+4c+4c+4c	5 5 1
13		"	"	4c+4c+8d	4 4 2
14		"	"	8d+8d	3 3 3
15		4b	4a+4c	4c+4c+4c+4c	5 5 —
16		"	"	4c+4c+8d	4 4 1
17		"	"	8d+8d	3 3 2
18		"	4c+4c	4a+4c+4c+4c	5 5 —
19		"	"	4a+4c+8d	4 4 1
20		"	"	4c+4c+4c+4c	6 6 —
21		"	"	4c+4c+8d	5 5 1
22		"	"	8d+8d	4 4 2
23		"	8d	4a+4c+4c+4c	4 4 1
24		"	"	4a+4c+8d	3 3 2
25		"	"	4c+4c+4c+4c	5 5 1
26		"	"	4c+4c+8d	4 4 2

TABLE 3. (Cont.)

Combination designation	Space Group	4 Fe	8 Sb	16 S	Number of Parameters		
					<i>x</i>	<i>y</i>	<i>z</i>
27	<i>Pnam</i> (V_h^{16})	"	$8d$	$8d+8d$	3	3	3
28		$4c$	$4a+4b$	$4c+4c+4c+4c$	5	5	—
29		"	"	$4c+4c+4d$	4	4	1
30		"	"	$8d+8d$	3	3	2
31		"	$4a+4c$	$4b+4c+4c+4c$	5	5	—
32		"	"	$4b+4c+8d$	4	4	1
33		"	"	$4c+4c+4c+4c$	6	6	—
34		"	"	$4c+4c+8d$	5	5	1
35		"	"	$8d+8d$	4	4	2
36		"	$4b+4c$	$4a+4c+4c+4c$	5	5	—
37		"	"	$4a+4c+8d$	4	4	1
38		"	"	$4c+4c+4c+4c$	6	6	—
39		"	"	$4c+4c+8d$	5	5	1
40		"	"	$8d+8d$	4	4	2
41		"	$4c+4c$	$4a+4b+4c+4c$	5	5	—
42		"	"	$4a+4b+8d$	4	4	1
43		$4c$	$4c+4c$	$4a+4c+4c+4c$	6	6	—
44		"	"	$4a+4c+8d$	5	5	1
45		"	"	$4b+4c+4c+4c$	6	6	—
46		"	"	$4b+4c+8d$	4	4	1
47		"	"	$4c+4c+4c+4c$	7	7	—
48		"	"	$4c+4c+8d$	6	6	1
49		"	"	$8d+8d$	5	5	2
50		"	$8d$	$4a+4b+4c+4c$	4	4	1
51		"	"	$4a+4b+8d$	3	3	2
52		"	"	$4a+4c+4c+4c$	5	5	1
53		"	"	$4a+4c+8d$	4	4	2
54		"	"	$4b+4c+4c+4c$	5	5	1
55		"	"	$4b+4c+8d$	4	4	2
56		"	"	$4c+4c+4c+4c$	6	6	1
57		"	"	$4c+4c+8d$	5	5	2
58		"	"	$8d+8d$	4	4	3

THE STRUCTURE AND CHEMICAL COMPOSITION OF GREENALITE

JOHN W. GRUNER, *University of Minnesota.*

INTRODUCTION

More than two years ago the writer made an x-ray examination of greenalite from the iron formation of the Mesabi Range in Minnesota. It was noticed that the powder of greenalite gave a definite pattern on which that of quartz was superimposed. The pattern did not resemble chlorite (or thüringite which is definitely a chlorite), nor biotite, nor glauconite or stilpnomelane. Neither was any resemblance observed between greenalite and chamosite, which gives a pattern different from any mentioned above. About a year ago the writer, when investigating serpentine, noticed a striking similarity in the powder photographs of it and greenalite. About this time Jolliffe¹ published a paper containing microscopic and chemical data on greenalite with which the present findings could not be reconciled.

One of the great difficulties is the securing of specimens of greenalite rock from which greenalite granules² can be separated. Three drill cores were finally found that answered the purpose. Also material identical with that used by Jolliffe was obtained through the kindness of Dr. C. K. Leith from the collection at Madison, Wisconsin. Three drill cores came from the following localities and depths:

No. 13156A, Depth about 550 ft. Section 24, T. 58 N., R. 17 W., near Gilbert.

No. 13156B, from the same drill hole but different, though unknown depth. Catalog No. M. 315S.

No. 11053, Depth about 905 ft. Section 35, T. 58 N., R. 18 W., near Virginia.

Dr. Leith's specimen came from a test pit near Biwabik. Generous grants from the Graduate School of the University of Minnesota have made this study possible. The writer is also indebted to Mr. Russel Wayland and Mr. Samuel Goldich for valuable assistance.

PHYSICAL AND CHEMICAL DATA

Microscopic examination of the drill cores confirmed largely the findings of Jolliffe in his single specimen. His "pure" isotropic greenalite has an index near 1.670, though many grains may be found which are below 1.655. Such differences may be due to sub-microscopic admixtures

¹ Jolliffe, Fred., A study of greenalite: *Am. Mineral.*, vol. 20, pp. 405-425, 1935.

² For the occurrence of greenalite and greenalite rock, the reader is referred to Jolliffe's paper. In thin sections of a single specimen of greenalite rock he found three significant minerals besides quartz. He named them greenalite, metagreenalite, and mineral X. The weight per cent. of these minerals based on micrometric analyses of six thin sections are 12.9, 34.6, and 44.2, respectively. These figures include matrix as well as granules.

of quartz. The writer is still of the same opinion, expressed ten years ago,³ that this greenalite is not isotropic when viewed with a very strong source of light. Jolliffe calls the material which is definitely anisotropic, metagreenalite, formed "through incipient crystallization" of greenalite. Obviously this distinction is one of degree of crystallization only. The finer grained the "metagreenalite" the more closely it would resemble "isotropic" greenalite.

Jolliffe's "mineral X" is of great interest. It has been mistaken for sericite, also for one of the amphiboles. It also resembles talc. Its optical properties do not fit any of these minerals completely, as Jolliffe has already pointed out. It might possibly be an iron serpentine, as will appear later. At any rate, its percentage in the different greenalite rock specimens is not as great as in that from Biwabik, described by Jolliffe. For example, in thin section No. 13156B, it is present in only very minor amounts, not exceeding 10 per cent. of the total volume. In No. 13156A it is practically absent. This is a very important fact for *x*-ray powder photographs of these samples are identical with those of other greenalite concentrates eliminating, therefore, mineral X as a source of the *x*-ray pattern recorded in Table 1.

The amount of quartz in greenalite rock can be estimated only very roughly, for some of it is extremely fine grained and only visible under high magnifications. Jolliffe seems to have greatly underestimated the quartz in his thin sections for he gives an average of only 5.7 per cent. by weight while his analysis shows 19.53 per cent. of "insoluble" SiO_2 .⁴ *X*-ray powder photographs show complete strong quartz patterns even after a bromoform separation.

To free greenalite from quartz the material was crushed to 100 or 150 mesh and placed in bromoform. More quartz was eliminated by running the material through a three stage magnetic separator. But the best method seems to be the dielectric one which also proved so successful with glauconite.⁵ All three methods were used in succession, but only in the sample furnished by Dr. Leith could the quartz be eliminated to such an extent that even its strongest *x*-ray diffraction lines did not show in the films. But about 5 per cent. of this sample was "mineral X," even after this treatment.

In Table 1 the *x*-ray pattern of greenalite is compared with those of antigorite, picrolite and precious serpentine. A striking resemblance is noted which is even more pronounced in the actual negatives. The in-

³ Gruner, J. W., Contributions to the geology of the Mesabi Range; *Minnesota Geol. Surv., Bull.* 19, p. 57, 1924.

⁴ *Op. cit.* Table 3 and p. 416.

⁵ Gruner, J. W., *Am. Mineral.*, vol. 20, p. 699, 1935.

dices which are given in the last column of the table are based on those published for chrysotile by Warren and Bragg.⁶ They agree closely. No other indices could be definitely identified, so far, for either serpentine or greenalite, but the writer believes that line No. 29 has the indices 0120 which would make the unit cell of greenalite slightly greater than that of serpentine along the *b*-axis.

No corrections were applied to the readings in Table. 1. Therefore, the interplanar distances *d* are a trifle too small in the upper part of the table. Not all lines have the same sharpness of definition. For example, Nos. 12 and 17 are broad and especially No. 17 is difficult to define. Lines sharp in serpentines are also relatively sharp in greenalite. The intensities also show good agreement when one considers that the serpentines x-rayed are very low in iron as compared with greenalite. This difference also explains the discrepancies in *d* values which are not any greater than those encountered, for example, in different members of the garnet group. Whether the β angle of the unit cell is also affected is impossible to tell. It may be assumed that the lines under the same number in Table 1 originate from corresponding planes, though this may not be true for all of them.

When Leith published his analyses of greenalite⁷ he assumed that the portion of the rock which was insoluble in HCl and in a 5 per cent. solution of Na₂CO₃ was quartz. This was more or less substantiated by microscopic examination of the residue. The analyses, exclusive of the insoluble SiO₂ and recalculated by Clark to 100 per cent., are given in Table 2. Jolliffe proceeded differently. He analyzed the rock⁸ and instead of subtracting his 19.53 per cent. insoluble SiO₂, deducted only 5.7 per cent. of quartz which he saw under the microscope. In other words, most of his insoluble SiO₂ was averaged in with his three iron silicates. The writer recalculated Jolliffe's analysis on the assumption that the insoluble SiO₂ was quartz and obtained the figures given in column 4 of Table 2.

Jolliffe made an analysis of his mineral X without stating, however, how he was able to separate this extremely fine grained fibrous mineral from the rest, after he admitted⁹ "that so many finely divided inclusions of other minerals are contained in the greenalite that its separation for chemical analysis is impossible." It seems to the writer the same statement applies to mineral X. Since nothing is said about soluble or insol-

⁶ Warren, B. E., and Bragg, W. L., The structure of chrysotile H₄Mg₃Si₂O₉: *Zeit. Krist.*, vol. 76, p. 201, 1931.

⁷ Leith, C. K., *U. S. Geol. Survey, Monograph* 43, p. 108, 1903.

⁸ *Op. cit.*, pp. 416 and 417.

⁹ *Op. cit.*, p. 408.

uble SiO_2 in this analysis¹⁰ it is difficult to accept his analysis as that of mineral X.

In order to ascertain whether greenalite contains about the same amount of Fe_2O_3 at depth as at the surface, Dr. R. Ellestad analyzed the iron in two of the drill cores for the writer with the following results.

	No. 13156A	No. 11053
FeO	24.35	31.08
Fe_2O_3	5.39	11.70

No. 11053 had been treated with bromoform. Therefore, the total percentage of iron is higher in it. Practically no iron oxides could be detected in these samples. Comparison with the analysis in Table 2 will show that the ratio of Fe_2O_3 to FeO in test pit material and deep drill cores is not materially different. Of great importance is the fact that No. 13156A, as seen in thin section, contains practically no "mineral X." Therefore, the Fe_2O_3 which Jolliffe¹¹ assigns to mineral X must be in the greenalite, in this specimen at least. Though no analysis was made for MgO in No. 13156A its presence in amounts proportional to those in other greenalite rocks is likely. Obviously, it would have to be in the greenalite and not in mineral X as Jolliffe proposes for greenalite rock in general.

DISCUSSION

The chemical evidence presented favors a composition for greenalite (including "metagreenalite") which is very similar to that proposed by Leith and Clark in 1903. The molecular ratios presented in Table 2 seem to agree well with a formula which given in oxide form is:¹²



The theoretical composition and molecular ratios of such a formula are recorded in the last column of Table 2. A serpentine whose magnesium was replaced by iron would correspond to this formula. Ordinarily iron in serpentine is largely ferrous but Doelter's *Handbuch der Mineralchemie*¹³ contains a number of reliable analyses in which Fe_2O_3 may be as high as 8 per cent.

The outstanding fact in the present investigation is that greenalite, when properly concentrated, gives an x-ray powder diagram which indicates that it is like serpentine in structure. Its unit cell when compared with serpentine gives almost the same dimensions for a_0 and b_0 , namely 14.5 Å and 18.6 Å, respectively. If c_0 corresponds to that of chrysotile,

¹⁰ *Op. cit.*, p. 416.

¹¹ *Op. cit.*, p. 423.

¹² The very high Fe_2O_3 content in the first two analyses of Table 2 seems to be partly due to iron oxide minerals in the two samples judging from Leith's description. *Op. cit.*, p. 109.

¹³ Vol. 2, 2nd Häft., p. 385.

the theoretical density of greenalite is 3.25, on the assumption that 2 molecules of $(\text{OH})_{12}\text{Fe}_9^{11}\text{Fe}_2^{11}\text{Si}_3\text{O}_{22}\cdot 2\text{H}_2\text{O}$ are contained in it. Since probably a few per cent. of magnesium always replace Fe^{11} the actual density would be less. The specific gravity of greenalite rock varies between 2.7 and 3.0, while the density of the granules lies between 2.85 and 3.15, according to Jolliffe. Since most of the impurities in the granules are considerably lower in gravity than greenalite itself the theoretical and measured densities seem to be in fair agreement.

The occurrence of greenalite is almost entirely confined to the Biwabik formation of the Mesabi Range in Minnesota. Such restriction cannot be accidental but must be due to very special conditions. The almost total absence of Ca, Al and alkalis in the solutions from which greenalite was precipitated colloiddally may have been a factor. Possibly relatively low concentration of CO_2 caused greenalite to form in certain layers in preference to iron carbonates and chert, which are preponderant in the iron formations of Michigan.

A serpentine structure with such an abundance of iron might be rather metastable, especially in its early stages of deposition. Slight changes in conditions might cause it to break down into iron oxides (mostly magnetite), siderite and quartz (usually called chert). The preservation of the granule texture of such layers might be the only remaining witness of the former existence of greenalite. The often described ferruginous cherts are of exactly such a nature. They are far more abundant than the greenalite layers which are interstratified with them, but are of the same granule texture.¹⁴

SUMMARY

Four samples of greenalite rock from the iron formation of the Mesabi Range were investigated microscopically, by x -rays and partly chemically. It was possible to separate greenalite from quartz, siderite and Jolliffe's mineral X by the combined applications of three methods, namely, bromoform solution, magnetic separation, and dielectric separation.

Greenalite is a distinct mineral species which has the crystal structure of such serpentines as antigorite and precious serpentine. These, however, are somewhat different in structure from the fibrous varieties. The chemical formula of greenalite approaches $9\text{FeO}\cdot\text{Fe}_2\text{O}_3\cdot 8\text{SiO}_2\cdot 8\text{H}_2\text{O}$, which structurally is $(\text{OH})_{12}\text{Fe}_9^{11}\text{Fe}_2^{11}\text{Si}_3\text{O}_{22}\cdot 2\text{H}_2\text{O}$. The ratio of Fe_2O_3 : FeO varies considerably, of course. Also, several per cent. of MgO usually replaces FeO . The theoretical density of an iron serpentine of this compo-

¹⁴ Gruner, J. W., Contribution to the geology of the Mesabi Range: *Minnesota Geol. Surv., Bull.* 19, pp. 10-17, 1924.

sition is 3.25. It is believed that greenalite appears to be isotropic because it is extremely fine grained and dark in color. Actually it is anisotropic like the metagreenalite of Jolliffe which is simply coarser grained greenalite. Jolliffe's formula $2\text{H}_2\text{O} \cdot 3\text{FeO} \cdot 4\text{SiO}_2$ for greenalite is too high in SiO_2 , because he based his SiO_2 ratio on the "soluble" and "insoluble" percentages of SiO_2 instead of discarding the insoluble portion as quartz as was done by Leith.

TABLE 1. COMPARISON OF POWDER PHOTOGRAPHS OF GREENALITE AND VARIOUS SERPENTINES. Fe RADIATION. RADIUS 57.3 MM.

Line	Greenalite ¹		Antigorite ²		Picrolite ³		Precious Serpentine ⁴		Indices
No.	<i>d</i>	<i>I</i>	<i>d</i>	<i>I</i>	<i>d</i>	<i>I</i>	<i>d</i>	<i>I</i>	
1	7.12	4	7.16	6	7.16	8	7.12	6	200
2			4.66	0.5	4.68	0.5	4.61	1	
3					4.22	0.5			
4	3.95	0.5	3.96	2	3.98	2	3.95	2	β 400
5					3.85	0.5	3.81	0.5	
6	3.559	4	3.588	7	3.592	9	3.592	7	400
7			3.48	0.5	3.462	0.5	3.490	0.5	
8	3.169	0.5							
9	3.010	0.5							
10	2.849	1	2.798	1	2.802	1	2.784	1	β of 12
11	2.737	1			2.671	0.5	2.677	0.5	
12	2.571	5	2.521	4	2.535	5	2.514	4	
13	2.457	1					2.450	0.5	
14	2.407	0.5	2.402	1	2.418	2	2.411	2	600
15					2.364	0.5	2.370	0.5	β of 17
16	2.334	0.5					2.210	0.5	
17	2.184	2	2.152	1	2.165	1	2.150	2	
18	2.058	0.5					2.012	0.5	
19	1.989	0.5	1.986	0.5	1.991	0.5	1.962	0.5	
20	1.928	0.5							
21	1.887	0.5					1.880	0.5	
22			1.848	0.5			1.831	1	
23	1.809	1	1.808	1	1.812	2	1.808	1	800
24					1.778	0.5	1.778	1	
25	1.759	0.5	1.723	0.5	1.729	1	1.722	1	β of 28
26	1.713	0.5	1.695	0.5	1.699	0.5	1.693	0.5	β of 29
27	1.647	0.5			1.592	0.5	1.584	0.5	
28	1.593	3	1.562	3	1.567	3	1.560	3	
29	1.553	2	1.538	2	1.540	2	1.536	2	
30			1.529	1	1.527	1	1.522	1	
31	1.509	1	1.509	1	1.509	1	1.503	1	
32							1.494	1	
33			1.478	0.5	1.471	0.5	1.472	0.5	
34	1.458	0.5	1.454	0.5			1.459	0.5	

TABLE 1. (Cont.)

Line	Greenalite ¹		Antigorite ²		Picrolite ³		Precious Serpentine ⁴		Indices
No.	<i>d</i>	<i>I</i>	<i>d</i>	<i>I</i>	<i>d</i>	<i>I</i>	<i>d</i>	<i>I</i>	
35			1.442	1	1.447	2	1.446	1	1000
36	1.428	0.5	1.417	0.5	1.416	0.5	1.410	0.5	
37	1.377	0.5					1.381	0.5	
38	1.354	0.5					1.358	0.5	
39			1.342	0.5			1.341	0.5	
40							1.328	0.5	
41	1.324	0.5	1.315	1	1.316	1	1.312	2	
42			1.298	0.5	1.296	0.5	1.295	0.5	
43	1.288	1					1.284	0.5	
44							1.273	0.5	
45			1.259	0.5			1.258	0.5	
46	1.246	0.5					1.244	0.5	
47	1.194	0.5	1.201	1	1.205	2	1.206	1	1200

¹ Dr. Leith's specimen.² Antigorio Valley, Italy.³ Chester, Mass.⁴ Montville, N. J.TABLE 2. ANALYSES OF GREENALITE ROCK EXCLUSIVE OF INSOLUBLE SiO₂

45758*			45765*		45766*		Jolliffe†		Iron Serpentine	
		Ratio		Ratio		Ratio		Ratio		Ratio
SiO ₂	30.08	.501	30.49	.508	38.00	.633	35.92	.598	33.58	.559
Fe ₂ O ₃	34.85	.218	23.52	.147	8.40	.052	9.80	.061	11.16	.070
FeO	25.72	.357	36.92	.513	46.56	.648	46.16	.643	45.19	.629
H ₂ O	9.35	.519	9.07	.503	7.04	.391	8.11	.450	10.07	.559
Total	100.00		100.00		100.00		99.99		100.00	

* Leith, C. K., *Op. cit.*, p. 246.† CO₂ was combined with a corresponding amount of FeO and deducted. MnO, MgO, and CaO were recalculated into their equivalents of FeO. Therefore, the treatment of this analysis corresponds to those of Leith's.

NOTES AND NEWS

POSSIBLE AGE OF MONAZITE FROM MARS HILL, NORTH CAROLINA*

JOHN PUTNAM MARBLE¹

The largest specimen of monazite thus far known was recently described by Schaller.² Through his kindness, portions of less perfect crystals accompanying the type specimen were given the writer for study. The mineral is reasonably fresh and unaltered in appearance, and the accompanying radiograph, Fig. 1, indicates a generally uniform distribution of radioactive material. The very low uranium content shown in the analyses is also to be taken as evidence that the degree of alteration is slight, according to Fenner.³



FIG. 1. Radiograph of representative crystal of Mars Hill, N. C., monazite.
1 week's exposure.

No attempt was made at a complete analysis, as the crystallographic and optical properties of the material were quite definite, and there are many monazite analyses available. The excellent analytical methods due to Fenner were used, slightly modified, with the following results.

TABLE 1

Sample in grams	PbSO ₄ , g.	≈ %Pb	Sample in grams	ThO ₂ , g.	≈ %Th
28.7857	0.0640	0.152	7.3428	0.4617	5.526
17.14108	0.04380	0.175	6.00088	0.38210	5.596

* Contribution from the Committee on the Determination of Geologic Time.

¹ Research Associate, Committee on the Determination of Geologic Time, Division of Geology and Geography, National Research Council.

² Schaller, W. T., *Am. Mineral.*, vol. 18, pp. 435-9, 1933.

³ Fenner, C. N., *Am. Jour. Sci.* [5] vol. 16, pp. 369-81, 1928.

Sample in grams	U ₃ O ₈ , g.	≈%U	Pb	
			U+0.36 Th	
6.9609	0.0016	0.019	0.076	Pb=207.90 Th=232.12
5.73249	0.00108	0.016	0.086	U=238.14

Assuming the absence of "ordinary" lead from this material we may calculate its age as 584 million years, using the formula:—

$$\frac{\log (U+0.36 \text{ Th}+1.155 \text{ Pb})-(\log U+0.36 \text{ Th})}{6.6 \times 10^{-5}} \text{ million years.}$$

It is hoped that the isotopic composition of the contained lead will be determined in the near future. Until this has been done, the above age is only an approximation. The field evidence would connect this monazite with the Cranberry Granite, presumably of pre-Cambrian age, a correlation in fair accord with the figure here found.

The courtesy of the Director of the United States Geological Survey and of the Assistant Secretary of the United States National Museum, for extending the facilities of their laboratories is gratefully acknowledged, as is the assistance of Dr. C. N. Fenner in the utilization of his analytical methods.

DICKITE FROM ST. LOUIS COUNTY, MISSOURI

VICTOR T. ALLEN, *St. Louis University, St. Louis, Mo.*

In a collection of geodes in the museum of St. Louis University, donated by V. Sosnovce in 1905, is one labeled "Flint Geode with Kaolin Endomorph, St. Louis County, Missouri." The writer has been aware for some time that the kaolin mineral in this geode is dickite, but postponed publishing a description of the occurrence until more exact information was available regarding the locality. Since the excellent paper by Tarr and Keller¹ on "Dickite in Missouri" has appeared it seems desirable to record this additional occurrence of dickite in Missouri even though the section of St. Louis county and the geologic formation furnishing the specimen are unknown. With the accepted hydrothermal origin of dickite, evidence is thus afforded that hydrothermal solutions reached east central Missouri during post-Mississippian time.

The geode (Fig. 1), which is approximately 4×6 centimeters, is composed of layers of chalcedony and quartz and encloses near the center a mass of dickite 8×18 millimeters, and near the side a smaller one 5×10 millimeters. No sulphides are associated with these minerals. The con-

¹ Tarr, W. A., and Keller, W. D., Dickite in Missouri: *Am. Mineral.*, vol. 21, pp. 109-114, 1936.

tact between the chalcedony and the dickite is sharp and lacks the leached porous zone mentioned by Tarr and Keller in the occurrence near Columbia.

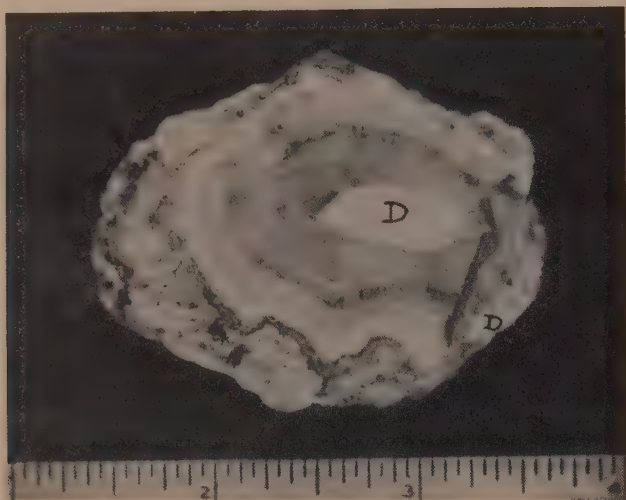


FIG. 1. Chalcedony and quartz geode enclosing dickite (D) near the center and at the side. About natural size. St. Louis Co., Mo.

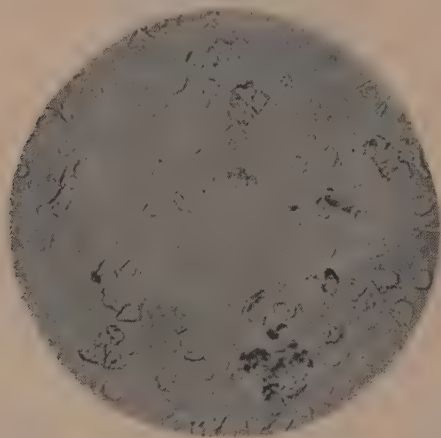


FIG. 2. Photomicrograph of dickite in geode from St. Louis Co., Mo. $\times 146$.

The dickite is snow white and has a distinctly glistening appearance. Under the microscope many of the cleavage flakes have the typical hexagonal shape of dickite (Fig. 2). The maximum diameter of the

crystals observed was .028 millimeters. The refractive index γ of the cleavage plates was $1.565 \pm .003$, and the maximum observed extinction angle on $\{010\}$ against the base was 18° .

AN OCCURRENCE OF LARGE ZIRCON NEEDLES
IN A BASIC PEGMATITE

RAY WILCOX, *University of Wisconsin.*

Crystals of zircon up to $7\frac{1}{4}$ inches long and $1/16$ to $1/8$ inch in diameter occur in a basic pegmatite in the SW $\frac{1}{4}$ of section 29, T 45 N, R 2 W, about 2 miles north of the town of Mellen, Wisconsin. The pegmatite, which is about $1\frac{1}{2}$ feet wide, cuts the gabbro country rock. It contains large crystals of basic plagioclase, hornblende and biotite, penetrated by the needles of zircon.

The zircon is non-magnetic, has a specific gravity greater than 3 and shows prismatic parting. Its color is cinnamon brown with adamantine lustre. The crystals are uniaxial and positive, showing parallel extinction. The refractive index of the ordinary ray is 1.925 ± 0.002 , and the birefringence determined on thin sections of grains with the universal stage is 0.054 ± 0.002 . An x-ray powder photograph by George W. Field of the University of Wisconsin shows a typical zircon pattern.

Dr. Henri Mngemach, the noted crystallographer and curator of the collections at Strasbourg, died on the night of June 10th at Strasbourg.

PROCEEDINGS OF SOCIETIES

MINERALOGICAL SOCIETY OF AMERICA

RESULTS OF BALLOT ON CONSTITUTIONAL AMENDMENTS AND CHANGES IN BY-LAWS

The Secretary's Office wishes to announce the results of voting on the proposed changes in the By-laws and the Constitutional Amendments recently submitted to the membership.

Ballots were returned as follows:

Members voting	107
Fellows voting	60

Tabulated results as given below indicate that all of the proposals submitted have been carried by a substantial majority.

PROPOSED CHANGES IN THE BY-LAWS

Article I—Membership

Section 1 . . . Any person or corporation interested in mineralogy, crystallography, *petrography* or allied sciences, shall be eligible to membership.

Yes 164 No 3

Section 2. Election (a) Fellows . . .

(b) Members. *Candidates for membership in the Mineralogical Society of America should be endorsed by at least one fellow of the Society and the application approved by either the secretary or treasurer acting for the Council.*

Yes 162 No 5

Article II—Dues

Section 1 . . . *The annual dues for fellows of the Mineralogical Society of America who also pay dues as fellows of the Geological Society of America shall be two dollars (\$2.00) payable at or before the annual meeting in advance. This provision shall continue in effect as long as the Geological Society of America shall contribute funds which the Council of the Mineralogical Society of America shall consider adequate to assist in publication of the Journal of the Mineralogical Society of America.*

Yes 166 No 1

Article V—Publications

The Society shall publish a journal devoted to the advancement of mineralogy, crystallography, *petrography*, and allied sciences.

Yes 164 No 3

PROPOSED AMENDMENTS TO THE CONSTITUTION

Article II—Object

The object of this Society shall be the advancement of mineralogy, crystallography, *petrography* and allied sciences.

Yes 58 No 2

Article IV—Membership

Section 2. Fellows shall be persons who have published results of research on mineralogy, crystallography, *petrography* or allied sciences, and who upon nomination by the Council shall have been duly elected to fellowship in the Society

Yes 58 No 2

Section 4. Patrons shall be persons who have bestowed important favors upon the Society. Election to patronship carries with it the rights and privileges of *fellows*.

Yes 59 No 1

Respectfully submitted,

PAUL F. KERR, *Secretary*

PHILADELPHIA MINERALOGICAL SOCIETY

Academy of Natural Sciences of Philadelphia, April 2, 1936

Mr. Arndt presided at a stated meeting of the Philadelphia Mineralogical Society, 44 members and 43 visitors being present.

Mr. O. Ivan Lee described "Mineral Collecting in North Carolina." Mr. Lee related in detail his experiences in collecting mineral specimens of the rare metals in western North Carolina. His talk was illustrated with lantern slides, photographs and specimens.

Other exhibits included; magnetite crystals in chlorite from Camp Horseshoe, Rising Sun, Md.; and spinel-twins of magnetite in chlorite from Miquon, Pa. Leonard A. Morgan reported calcite and pink dolomite from East Petersburg, Pa., and trilobites from Prudenville, near Lancaster, Pa.

W. H. FLACK, *Secretary*

NEW HAVEN MINERAL CLUB

At the Annual Meeting of the New Haven Mineral Club, held in October, the following officers were elected:

President: Arthur Sandiford

Vice-President: William H. Otersen

Treasurer: Charles Thomas

Secretary: Frank Wilson

The club has enjoyed an unusually fine winter season with the following speakers and their subjects.

November, Charles Thomas: "Mineral Collecting in Connecticut" with a fine representative display of minerals collected in Connecticut over a period of years.

December, Herman Levy: "Fossils with relation to minerals," accompanied by lantern slides and specimens.

January, Arthur Sandiford: "Micro Chemistry." A lecture with the aid of several microscopes that enabled most of the members to examine the specimens.

February, F. Fowler: "Luray Caverns."

March, Rufus S. Hart: "Iron Industry of Connecticut."

April, H. Thorpe: "Orio Dents Fossils."

The Club held its May meeting at Portland, Connecticut. It has been decided to discontinue the July and August outings due to hot weather and vacations.

CLEVELAND MINERALOGICAL SOCIETY

The Cleveland Mineralogical Society, since its organization on July 24, 1935, has been instrumental in bringing together persons having an interest in mineralogy, geology, and gemmology. The Society has an active enrollment of twenty-five members including residents of Cleveland and its suburbs, and others living in outlying towns and cities as distant as Youngstown. More than half of the group are engaged in academic or commercial professions which put them in contact, daily, with minerals or ores.

From a mineralogical standpoint, the Cleveland area is relatively barren. Many underlying strata of sedimentary deposits topped with glacial till have little to yield to the mineral hunter. In spite of this a survey by the organization reveals an abundance of the following: halite, recrystallized from the brine pumped from salt wells in the center of this city; nodules and spheres of well-crystallized pyrite, imbedded in carbonaceous shale along Euclid Creek in Cleveland Heights; thin tabular and twinned selenite crystals, occur in blue clay in cliffs on the shore of Lake Erie at Euclid; selenite rosettes, and perfect star-shaped penetration twins measuring two inches or less, found in glacial rubble near Kirtland, Ohio (massive selenite also reported to be found at Kirtland); crystals of selenite and penetration twins, in blue clay south of Ellesworth, Ohio, and at Canfield, Ohio; sphalerite in nodules incrustated with ferruginous shale (some of these nodules weigh several pounds), in creek near selenite deposit at Ellesworth, Ohio; chert, chalcedony, agate and other siliceous minerals at Flint Ridge, Ohio; massive gypsum and anhydrite, at Gypsum, Ohio; large scalenohedrons of calcite, in dolomite on the shore of Lake Erie at Catawba Island, Ohio; celestite, large clear crystals in caves on Strontian Island in Lake Erie; celestite, fluorescent fluorite (cubes), pyrite, and sphalerite, all found at Clay Center, Ohio.

Some of the above locations are on the Society's schedule for field trips this summer.

At the April meeting of the Society the following officers were elected:

PRESIDENT: George Whitaker

VICE-PRESIDENT: Gerald U. Greene

COUNCILLOR: A. S. Furcron

SECRETARY-TREASURER: Robert H. Long